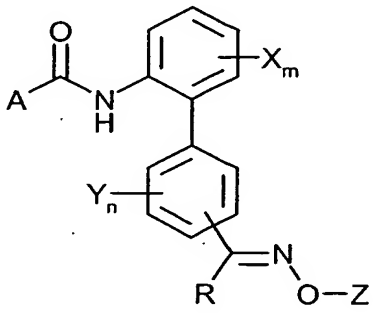
JC20 Rec'd PCT/PTO 07 JUN 2005

Biphenylcarboxamides

The present invention relates to novel biphenylcarboxamides, to a plurality of processes for their preparation and to their use for controlling unwanted microorganisms.

It is already known that numerous carboxanilides have fungicidal properties (compare WO 93/11 117, WO 99/09 013, WO 00/14 071, EP-A 0 545 099 and EP-A 0 589 301). In particular, WO 02/08195 and WO 02/08197 disclose oxyiminomethyl-substituted carboxamides. The activity of these compounds is good; however, it is sometimes unsatisfactory.

This invention now provides novel biphenylcarboxamides of the formula (I)



(I),

in which

R represents hydrogen, C_1-C_6-alkyl or C_1-C_3-haloalkyl having in each case 1 to 7 fluorine, chlorine and/or bromine atoms,

Z represents C_3-C_8-alkenyl, C_3-C_8-alkynyl, C_3-C_8-haloalkenyl, C_3-C_8-haloalkynyl having in each case 1 to 5 fluorine, chlorine and/or bromine atoms, or (C_3-C_8-cycloalkyl)(C_1-C_4-alkyl),

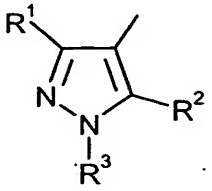
X and Y independently of one another represent halogen, cyano, nitro, C_1-C_8-alkyl, C_1-C_8-alkoxy, C_1-C_8-alkylthio, C_1-C_6-haloalkyl, C_1-C_6-haloalkoxy or C_1-C_6-haloalkylthio having in each case 1 to 13 fluorine, chlorine and/or bromine atoms,

m represents 0, 1, 2, 3 or 4, where x represents identical or different radicals if m represents 2, 3 or 4,

n represents 0, 1, 2, 3 or 4, where y represents identical or different radicals if n represents 2, 3 or 4,

and

A represents a radical of the formula



in which

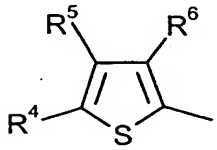
R^1 represents hydrogen, cyano, halogen, nitro, C_1-C_4-alkyl, C_3-C_6-cycloalkyl, C_1-C_4-alkoxy, C_1-C_4-alkylthio, aminocarbonyl, aminocarbonyl-C_1-C_4-alkyl or represents C_1-C_4-haloalkyl, C_1-C_4-haloalkoxy, C_1-C_4-haloalkylthio having in each case 1 to 5 halogen atoms, and

R^2 represents hydrogen, halogen, cyano, C_1-C_4-alkyl, C_1-C_4-alkoxy or C_1-C_4-alkylthio and

R^3 represents hydrogen, C_1-C_4-alkyl, hydroxy-C_1-C_4-alkyl, C_2-C_6-alkenyl, C_3-C_6-cycloalkyl, C_1-C_4-alkylthio-C_1-C_4-alkyl, C_1-C_4-alkoxy-C_1-C_4-alkyl, or represents C_1-C_4-haloalkyl, halo(C_1-C_4-alkylthio-C_1-C_4-alkyl), halo(C_1-C_4-alkoxy-C_1-C_4-alkyl) having in each case 1 to 5 halogen atoms or represents phenyl,

or

A represents a radical of the formula



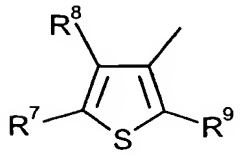
in which

R^4 and R^5 independently of one another represent hydrogen, halogen, C_1-C_4-alkyl or C_1-C_4-haloalkyl having 1 to 5 halogen atoms and

R^6 represents halogen, cyano or C_1-C_4-alkyl, C_1-C_4-haloalkyl, C_1-C_4-haloalkoxy having in each case 1 to 5 halogen atoms,

or

A represents a radical of the formula



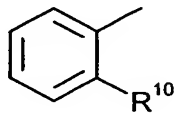
in which

R^7 and R^8 independently of one another represent hydrogen, halogen, C_1-C_4-alkyl or C_1-C_4-haloalkyl having 1 to 5 halogen atoms and

R^9 represents hydrogen, halogen or C_1-C_4-alkyl,

or

A represents a radical of the formula

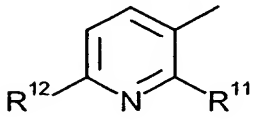


in which

R^{10} represents hydrogen, halogen, hydroxyl, cyano, C_1-C_6-alkyl, C_1-C_4-haloalkyl, C_1-C_4-haloalkoxy, C_1-C_4-haloalkylthio having in each case 1 to 5 halogen atoms,

or

A represents a radical of the formula



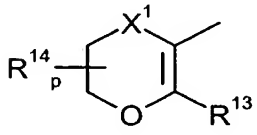
in which

R^{11} represents halogen, hydroxyl, cyano, C_1-C_4-alkyl, C_1-C_4-alkoxy, C_1-C_4-alkylthio, C_1-C_4-haloalkyl, C_1-C_4-haloalkoxy, C_1-C_4-haloalkylthio having in each case 1 to 5 halogen atoms and

R^{12} represents hydrogen, halogen, cyano, C_1-C_4-alkyl, C_1-C_4-alkoxy, C_1-C_4-alkylthio, C_1-C_4-alkylsulfinyl, C_1-C_4-alkylsulfonyl or represents C_1-C_4-haloalkyl, C_1-C_4-haloalkoxy having in each case 1 to 5 halogen atoms,

or

A represents a radical of the formula



in which

R^{13} represents C_1-C_4-alkyl or represents C_1-C_4-haloalkyl having 1 to 5 halogen atoms,

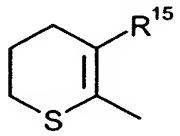
R^{14} represents C_1-C_4-alkyl,

X^1 represents S (sulfur), represents SO, SO_2 or CH_2 and

p represents 0, 1 or 2,

or

A represents a radical of the formula

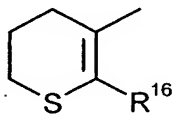


in which

R^{15} represents C_1-C_4-alkyl or represents C_1-C_4-haloalkyl having 1 to 5 halogen atoms,

or

A represents a radical of the formula

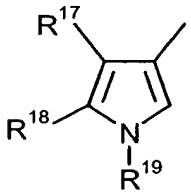


in which

R^{16} represents C_1-C_4-alkyl or represents C_1-C_4-haloalkyl having 1 to 5 halogen atoms,

or

A represents a radical of the formula



in which

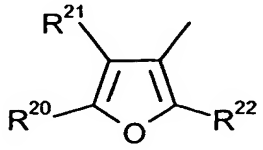
R^{17} represents halogen, cyano, C_1-C_4-alkyl or represents C_1-C_4-haloalkyl having 1 to 5 halogen atoms,

R^{18} represents hydrogen, halogen, C_1-C_4-alkyl or represents C_1-C_4-haloalkyl having 1 to 5 halogen atoms,

R^{19} represents hydrogen, cyano, C_1-C_4-alkyl, C_1-C_4-haloalkyl having 1 to 5 halogen atoms, C_1-C_4-alkoxy-C_1-C_4-alkyl, hydroxy-C_1-C_4-alkyl, C_1-C_4-alkysulfonyl, di(C_1-C_4-alkyl)aminosulfonyl, C_1-C_6-alkylcarbonyl or represents optionally substituted phenylsulfonyl or benzoyl,

or

A represents a radical of the formula



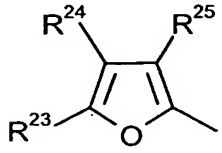
in which

R^{20} and R^{21} independently of one another represent hydrogen, halogen, amino, C_1-C_4-alkyl or represent C_1-C_4-haloalkyl having 1 to 5 halogen atoms and

R^{22} represents hydrogen, halogen, C_1-C_4-alkyl or represents C_1-C_4-haloalkyl having 1 to 5 halogen atoms,

or

A represents a radical of the formula



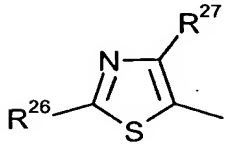
in which

R^{23} and R^{24} independently of one another represent hydrogen, halogen, amino, nitro, C_1-C_4-alkyl or represent C_1-C_4-haloalkyl having 1 to 5 halogen atoms and

R^{25} represents hydrogen, halogen, C_1-C_4-alkyl or represents C_1-C_4-haloalkyl having 1 to 5 halogen atoms,

or

A represents a radical of the formula



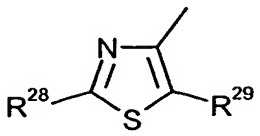
in which

R^{26} represents hydrogen, halogen, amino, C_1-C_4-alkylamino, di-(C_1-C_4-alkyl)amino, cyano, C_1-C_4-alkyl or represents C_1-C_4-haloalkyl having 1 to 5 halogen atoms and

R^{27} represents halogen, C_1-C_4-alkyl or C_1-C_4-haloalkyl having 1 to 5 halogen atoms,

or

A represents a radical of the formula



in which

R^{28} represents hydrogen, halogen, amino, C_1-C_4-alkylamino, di-(C_1-C_4-alkyl)amino, cyano, C_1-C_4-alkyl or represents C_1-C_4-haloalkyl having 1 to 5 halogen atoms and

R^{29} represents halogen, C_1-C_4-alkyl or C_1-C_4-haloalkyl having 1 to 5 halogen atoms,

or

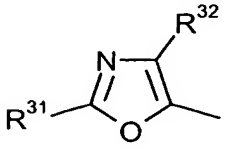
A represents a radical of the formula

in which

R^{30} represents halogen, C_1-C_4-alkyl, C_1-C_4-haloalkyl having 1 to 5 halogen atoms,

or

A represents a radical of the formula



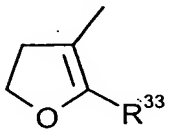
in which

R^{31} represents hydrogen or C_1-C_4-alkyl and

R^{32} represents halogen or C_1-C_4-alkyl,

or

A represents a radical of the formula

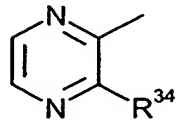


in which

R^{33} represents C_1-C_4-alkyl or C_1-C_4-haloalkyl having 1 to 5 halogen atoms,

or

A represents a radical of the formula



in which

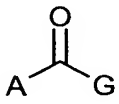
R^{34} represents hydrogen, halogen, C_1-C_4-alkyl or C_1-C_2-haloalkyl having 1 to 5 halogen atoms.

If appropriate, the compounds according to the invention can be present as mixtures of different possible isomeric forms, in particular of stereoisomers, such as, for example, E and Z, threo and erythro, and also optical isomers, and, if appropriate, also of tautomers. What is claimed are both the E and the Z isomers, and the threo and erytho-, and also the optical isomers, any mixtures of these isomers, and also the possible tautomeric forms.

In particular with reference to the oxime double bond, E/Z isomers occur when R is C_1-C_6-alkyl. These isomers are generally present as a mixture. The configuration given below in structural formulae always includes both possibilities. For the sake of simplicity, only one isomer is shown in each case.

Furthermore, it has been found that biphenylcarboxamides of the formula (I) are obtained when

a) carboxylic acid derivatives of the formula (II)



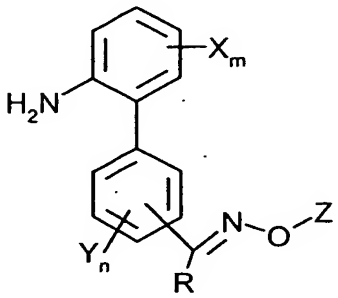
(II)

in which

A is as defined above and

G represents halogen, hydroxyl or C_1-C_6-alkoxy

are reacted with aniline derivatives of the formula (III)



(III)

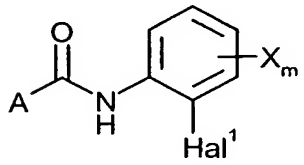
in which

R, Z, X, Y, m and n are as defined above,

if appropriate in the presence of a catalyst, if appropriate in the presence of an acid binder and if appropriate in the presence of a diluent,

or

b) carboxamide derivatives of the formula (IV)



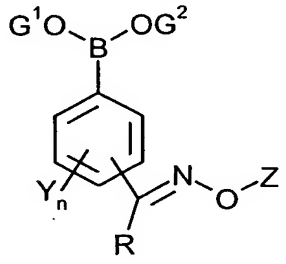
(IV)

in which

A, X and m are as defined above,

Hal^1 represents bromine or iodine,

are reacted with boronic acid derivatives of the formula (V)



(V)

in which

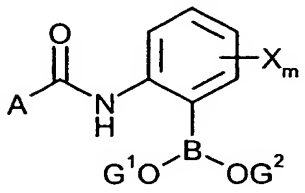
R, Z, Y and n are as defined above and

G^1 and G^2 each represent hydrogen or together represent tetramethylethylene

in the presence of a catalyst, if appropriate in the presence of an acid binder and if appropriate in the presence of a diluent,

or

c) carboxamide boronic acid derivatives of the formula (VI)



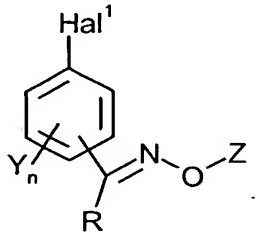
(VI)

in which

A, X and m are as defined above and

G^1 and G^2 each represent hydrogen or together represent tetramethylethylene

are reacted with phenyl oxime derivatives of the formula (VII)



(VII)

in which

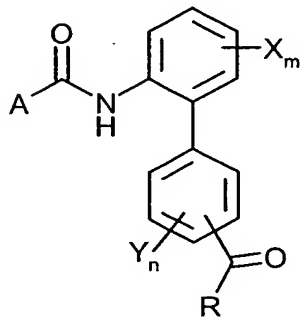
R, Z, Y and n are as defined above,

Hal^1 represents bromine or iodine,

in the presence of a catalyst, if appropriate in the presence of an acid binder and if appropriate in the presence of a diluent,

or

d) biphenylacyl derivatives of the formula (VIII)



(VIII)

in which

A, R, X, Y, m and n are as defined above,

are reacted with hydroxylamine derivatives of the formula (IX)

$$Z-O-NH_2 \times HCl \qquad (IX)$$

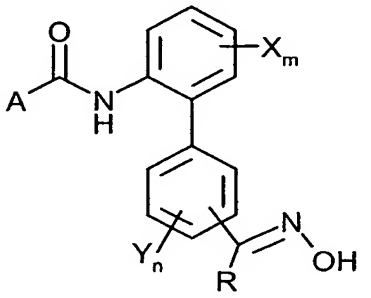
in which

Z is as defined above,

if appropriate in the presence of an acid binder and if appropriate in the presence of a diluent,

or

e) hydroxyimino derivatives of the formula (I-a)



(I-a)

in which

A, R, X, Y, m and n are as defined above

are reacted with compounds of the formula (X)

$$Z-E \qquad (X)$$

in which

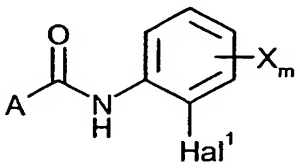
Z is as defined above,

E represents chlorine, bromine, iodine, methanesulfonyl or p-toluenesulfonyl,

if appropriate in the presence of an acid binder and if appropriate in the presence of a diluent,

or

f) carboxamide derivatives of the formula (IV)



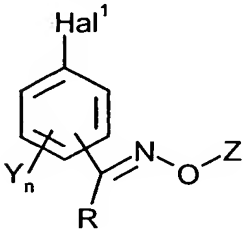
(IV)

in which

A, X and m are as defined above,

Hal^1 represents bromine or iodine,

are reacted with phenyl oxime derivatives of the formula (VII)



(VII)

in which

R, Z, Y and n are as defined above,

Hal^1 represents bromine or iodine

in the presence of a palladium or platinum catalyst and in the presence of 4,4,4',4',5,5,5',5'-octamethyl-2,2'-bis-1,3,2-dioxaborolane, if appropriate in the presence of an acid binder and if appropriate in the presence of a diluent.

Finally, it has been found that novel biphenylcarboxamides of the formula (I) have very good microbicidal properties and can be used for controlling unwanted microorganisms both in crop protection and in the protection of materials.

Surprisingly, the biphenylcarboxmides of the formula (I) according to the invention have considerably better fungicidal activity than the constitutionally most similar active compounds of the prior art having the same direction of action.

The formula (I) provides a general definition of the biphenylcarboxamides according to the invention.

R preferably represents hydrogen, C_1-C_4-alkyl or C_1-C_3-haloalkyl having in each case 1 to 7 fluorine, chlorine and/or bromine atoms.

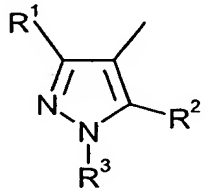
Z preferably represents C_3-C_6-alkenyl, C_3-C_6-alkynyl, C_3-C_6-haloalkenyl, C_3-C_6-haloalkynyl having in each case 1 to 5 fluorine, chlorine and/or bromine atoms, or (C_3-C_6-cycloalkyl)-(C_1-C_4-alkyl).

X and Y independently of one another preferably represent fluorine, chlorine, bromine, cyano, nitro, C_1-C_6-alkyl, C_1-C_6-alkoxy, C_1-C_6-alkylthio, C_1-C_2-haloalkyl, C_1-C_2-haloalkoxy or C_1-C_2-haloalkylthio having in each case 1 to 5 fluorine, chlorine and/or bromine atoms.

m preferably represents 0, 1, 2 or 3, where x represents identical or different radicals if m represents 2 or 3.

n preferably represents 0, 1, 2 or 3, where y represents identical or different radicals if n represents 2 or 3.

A preferably represents a radical of the formula



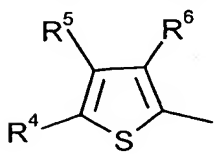
in which

R^1 represents hydrogen, cyano, fluorine, chlorine, bromine, iodine, methyl, ethyl, isopropyl, cyclopropyl, methoxy, ethoxy, methylthio, ethylthio, aminocarbonyl, aminocarbonylmethyl, aminocarbonylethyl, C_1-C_2-haloalkyl C_1-C_2-haloalkoxy having in each case 1 to 5 fluorine, chlorine and/or bromine atoms, trifluoromethylthio or difluoromethylthio,

R^2 represents hydrogen, fluorine, chlorine, bromine, iodine, methyl, ethyl, methoxy, ethoxy, methylthio or ethylthio and

R^3 represents hydrogen, methyl, ethyl, n-propyl, isopropyl, hydroxymethyl, hydroxyethyl, cyclopropyl, cyclopentyl, cyclohexyl, C_1-C_2-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms or represents phenyl.

A furthermore preferably represents a radical of the formula

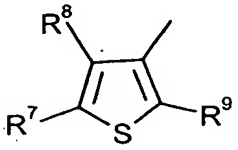


in which

R^4 and R^5 independently of one another represent hydrogen, fluorine, chlorine, bromine, methyl, ethyl or C_1-C_2-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms and

R^6 represents fluorine, chlorine, bromine, iodine, cyano, methyl, ethyl, trifluoromethyl or C_1-C_2-haloalkoxy having 1 to 5 fluorine, chlorine and/or bromine atoms.

A furthermore preferably represents a radical of the formula

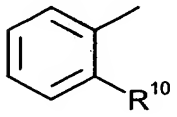


in which

R^7 and R^8 independently of one another represent hydrogen, fluorine, chlorine, bromine, methyl, ethyl or C_1-C_2-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms and

R^9 represents hydrogen, fluorine, chlorine, bromine, methyl or ethyl.

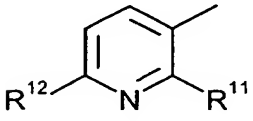
A furthermore preferably represents a radical of the formula



in which

R^{10} represents hydrogen, fluorine, chlorine, bromine, iodine, hydroxyl, cyano, C_1-C_4-alkyl, C_1-C_2-haloalkyl, C_1-C_2-haloalkoxy, C_1-C_2-haloalkylthio having in each case 1 to 5 fluorine, chlorine and/or bromine atoms.

A furthermore preferably represents a radical of the formula



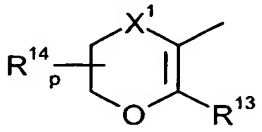
in which

R^{11} represents fluorine, chlorine, bromine, iodine, hydroxyl, cyano, C_1-C_4-alkyl, methoxy, ethoxy, methylthio, ethylthio, C_1-C_2-haloalkyl, C_1-C_2-haloalkoxy

having in each case 1 to 5 fluorine, chlorine and/or bromine atoms, trifluoromethylthio, difluoromethylthio and

R^{12} represents hydrogen, fluorine, chlorine, bromine, iodine, cyano, C_1-C_4-alkyl, methoxy, ethoxy, methylthio, ethylthio, C_1-C_2-alkylsulfinyl, C_1-C_2-alkylsulfonyl, C_1-C_2-haloalkyl, C_1-C_2-haloalkoxy having in each case 1 to 5 fluorine, chlorine and/or bromine atoms.

A furthermore preferably represents a radical of the formula



in which

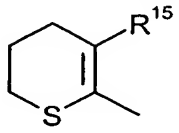
R^{13} represents methyl, ethyl or represents C_1-C_2-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms and

R^{14} represents methyl or ethyl,

X^1 represents S (sulfur), represents SO, SO_2 or CH_2 and

p represents 0, 1 or 2.

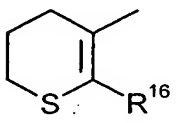
A furthermore preferably represents a radical of the formula



in which

R^{15} represents methyl, ethyl or represents C_1-C_2-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

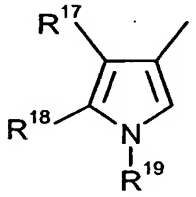
A furthermore preferably represents a radical of the formula



in which

R^{16} represents methyl, ethyl or represents C_1-C_2-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

A furthermore preferably represents a radical of the formula



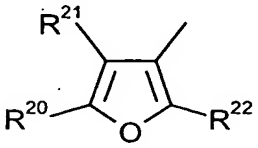
in which

R^{17} represents fluorine, chlorine, bromine, cyano, methyl, ethyl, isopropyl or represents C_1-C_2-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms,

R^{18} represents hydrogen, fluorine, chlorine, bromine, methyl, ethyl or represents C_1-C_2-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms and

R^{19} represents hydrogen, methyl, ethyl, C_1-C_2-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms, C_1-C_2-alkoxy-C_1-C_2-alkyl, hydroxymethyl, hydroxyethyl, methylsulfonyl or dimethylaminosulfonyl.

A furthermore preferably represents a radical of the formula

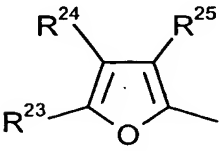


in which

R^{20} and R^{21} independently of one another represent hydrogen, fluorine, chlorine, bromine, amino, methyl, ethyl or represents C_1-C_2-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms and

R^{22} represents hydrogen, fluorine, chlorine, bromine, methyl, ethyl or represents C_1-C_2-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

A furthermore preferably represents a radical of the formula

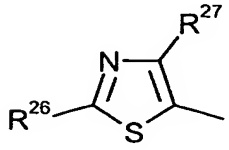


in which

R^{23} and R^{24} independently of one another represent hydrogen, fluorine, chlorine, bromine, amino, nitro, methyl, ethyl or represent C_1-C_2-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms and

R^{25} represents hydrogen, fluorine, chlorine, bromine, methyl, ethyl or represents C_1-C_2-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

A furthermore preferably represents a radical of the formula

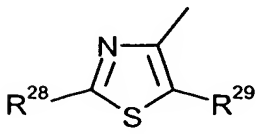


in which

R^{26} represents hydrogen, fluorine, chlorine, bromine, amino, C_1-C_4-alkylamino, di-(C_1-C_4-alkyl)amino, cyano, methyl, ethyl or represents C_1-C_2-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms and

R^{27} represents fluorine, chlorine, bromine, methyl, ethyl or C_1-C_2-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

A furthermore preferably represents a radical of the formula

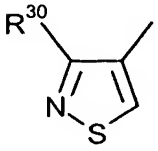


in which

R^{28} represents hydrogen, fluorine, chlorine, bromine, amino, C_1-C_4-alkylamino, di-(C_1-C_4-alkyl)amino, cyano, methyl, ethyl or represents C_1-C_2-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms and

R^{29} represents fluorine, chlorine, bromine, methyl, ethyl or C_1-C_2-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

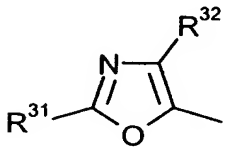
A furthermore preferably represents a radical of the formula



in which

R^{30} represents fluorine, chlorine, bromine, methyl, ethyl or C_1-C_2-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

A furthermore preferably represents a radical of the formula

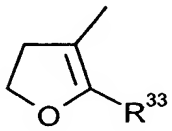


in which

R^{31} represents hydrogen, methyl or ethyl and

R^{32} represents fluorine, chlorine, bromine, methyl or ethyl.

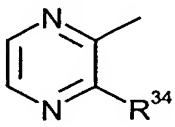
A furthermore preferably represents a radical of the formula



in which

R^{33} represents methyl, ethyl or C_1-C_2-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

A furthermore preferably represents a radical of the formula



in which

R^{34} represents hydrogen, fluorine, chlorine, bromine, methyl, ethyl or trifluoromethyl.

R particularly preferably represents hydrogen, methyl, ethyl, isopropyl, tert-butyl.

Z particularly preferably represents allyl, 2-butenyl, 2-methylallyl, 1-methylallyl, 3-methyl-2-butenyl, propargyl, 2-butynyl, 3-butynyl, 2-methyl-3-butynyl, 3,3-

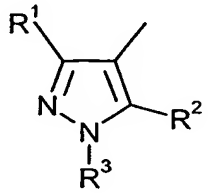
difluoroallyl, 3,3-dichloroallyl, cyclopropylmethyl, cyclopentylmethyl, cyclohexylmethyl.

X and Y independently of one another particularly preferably represent fluorine, chlorine, bromine, cyano, nitro, methyl, ethyl, n-propyl, isopropyl, n-butyl, sec-butyl, isobutyl, tert-butyl, methoxy, ethoxy, methylthio, trichloromethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl, difluoromethoxy, trifluoromethoxy, trifluoromethylthio, difluorochloromethylthio,

m particularly preferably represents 0 or 1.

n particularly preferably represents 0, 1 or 2, where y represents identical or different radicals if n represents 2.

A particularly preferably represents a radical of the formula



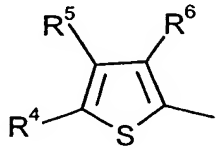
in which

R^1 represents hydrogen, fluorine, chlorine, bromine, iodine, methyl, ethyl, isopropyl, cyclopropyl, methoxy, ethoxy, methylthio, ethylthio, monofluoromethyl, difluoromethyl, trifluoromethyl, difluorochloromethyl, trichloromethyl, trifluoromethoxy, trichloromethoxy, trifluoromethylthio or difluoromethylthio and

R^2 represents hydrogen, fluorine, chlorine, bromine, iodine, methyl, ethyl, methoxy, ethoxy, methylthio or ethylthio and

R^3 represents hydrogen, methyl, ethyl, hydroxymethyl, hydroxyethyl, trifluoromethyl, difluoromethyl or phenyl.

A furthermore particularly preferably represents a radical of the formula

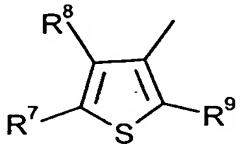


in which

R^4 and R^5 independently of one another represent hydrogen, fluorine, chlorine, bromine, methyl, ethyl, difluoromethyl, trifluoromethyl, difluorochloromethyl or trichloromethyl and

R^6 represents fluorine, chlorine, bromine, cyano, methyl, trifluoromethyl, trifluoromethoxy, difluoromethoxy, difluorochloromethoxy or trichloromethoxy.

A furthermore particularly preferably represents a radical of the formula

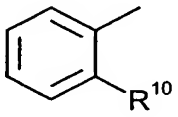


in which

R^7 and R^8 independently of one another represent hydrogen, fluorine, chlorine, bromine, methyl, ethyl, difluoromethyl, trifluoromethyl, difluorochloromethyl or trichloromethyl and

R^9 represents hydrogen, fluorine, chlorine, bromine, methyl or ethyl.

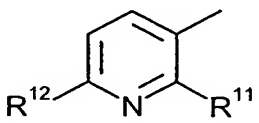
A furthermore particularly preferably represents a radical of the formula



in which

R^{10} represents hydrogen, fluorine, chlorine, bromine, iodine, hydroxyl, cyano, methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, difluoromethyl, trifluoromethyl, difluorochloromethyl, trichloromethyl, trifluoromethoxy, difluoromethoxy, difluorochloromethoxy, trichloromethoxy, trifluoromethylthio, difluoromethylthio, difluorochloromethylthio or trichloromethylthio.

A furthermore particularly preferably represents a radical of the formula

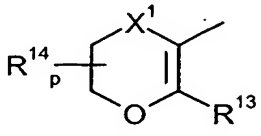


in which

R^{11} represents fluorine, chlorine, bromine, iodine, hydroxyl, cyano, methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, methoxy, ethoxy, methylthio, ethylthio, trifluoromethyl, difluoromethyl, difluorochloromethyl, trichloromethyl, trifluoromethoxy, difluoromethoxy, difluorochloromethoxy, trichloromethoxy, difluoromethylthio, trifluoromethylthio and

R^{12} represents hydrogen, fluorine, chlorine, bromine, iodine, cyano, methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, methoxy, ethoxy, methylthio, ethylthio, methylsulfinyl, methylsulfonyl, trifluoromethyl, difluoromethyl, difluorochloromethyl, trichloromethyl, trifluoromethoxy, difluoromethoxy, difluorochloromethoxy or trichloromethoxy.

A furthermore particularly preferably represents a radical of the formula



in which

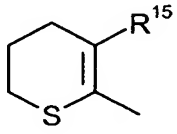
R^{13} represents methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl and

R^{14} represents methyl or ethyl,

X^1 represents S (sulfur), represents SO, SO_2 or CH_2 and

p represents 0, 1 or 2.

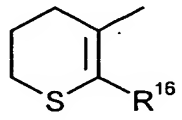
A furthermore particularly preferably represents a radical of the formula



in which

R^{15} represents methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.

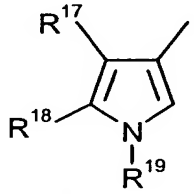
A furthermore particularly preferably represents a radical of the formula



in which

R^{16} represents methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.

A furthermore particularly preferably represents a radical of the formula



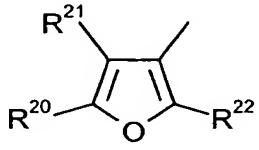
in which

R^{17} represents fluorine, chlorine, bromine, cyano, methyl, ethyl, isopropyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl,

R^{18} represents hydrogen, fluorine, chlorine, bromine, methyl, ethyl, trifluoromethyl, difluoromethyl or trichloromethyl and

R^{19} represents hydrogen, methyl, ethyl, trifluoromethyl, methoxymethyl, ethoxymethyl, hydroxymethyl or hydroxyethyl.

A furthermore particularly preferably represents a radical of the formula

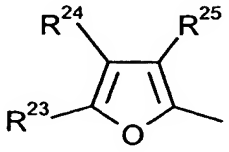


in which

R^{20} and R^{21} independently of one another represent hydrogen, fluorine, chlorine, bromine, methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl and

R^{22} represents hydrogen, fluorine, chlorine, bromine, methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.

A furthermore particularly preferably represents a radical of the formula

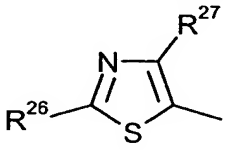


in which

R^{23} and R^{24} independently of one another represent hydrogen, fluorine, chlorine, bromine, nitro, methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl and

R^{25} represents hydrogen, fluorine, chlorine, bromine, methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.

A furthermore particularly preferably represents a radical of the formula

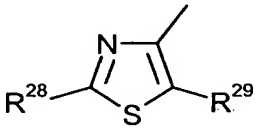


in which

R^{26} represents hydrogen, fluorine, chlorine, bromine, amino, methylamino, dimethylamino, cyano, methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl and

R^{27} represents fluorine, chlorine, bromine, methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.

A furthermore particularly preferably represents a radical of the formula

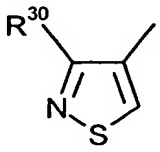


in which

R^{28} represents hydrogen, fluorine, chlorine, bromine, amino, methylamino, dimethylamino, cyano, methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl and

R^{29} represents fluorine, chlorine, bromine, methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.

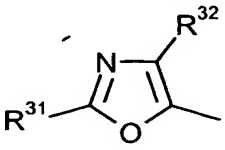
A furthermore particularly preferably represents a radical of the formula



in which

R^{30} represents fluorine, chlorine, bromine, methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.

A furthermore particularly preferably represents a radical of the formula

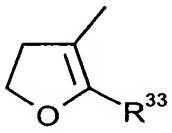


in which

R^{31} represents hydrogen, methyl or ethyl and

R^{32} represents fluorine, chlorine, bromine, methyl or ethyl.

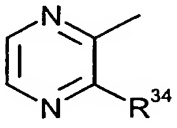
A furthermore particularly preferably represents a radical of the formula



in which

R^{33} represents methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.

A furthermore particularly preferably represents a radical of the formula



in which

R^{34} represents hydrogen, fluorine, chlorine, bromine, methyl, ethyl or trifluoromethyl.

R very particularly preferably represents hydrogen or methyl.

Z very particularly preferably represents allyl, 2-butenyl, 2-methylallyl, 1-methylallyl, 3-methyl-2-butenyl, propargyl, 2-butynyl, 2-methyl-3-butynyl, 3,3-difluoroallyl, cyclopropylmethyl.

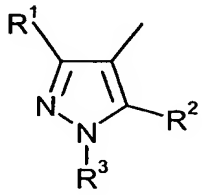
X very particularly preferably represents fluorine or methyl.

Y very particularly preferably represents fluorine, chlorine, bromine, cyano, methyl, methoxy, methylthio, trichloromethyl, trifluoromethyl, trifluoromethoxy, trifluoromethylthio.

m very particularly preferably represents 0 or 1.

n very particularly preferably represents 0 or 1.

A very particularly preferably represents a radical of the formula



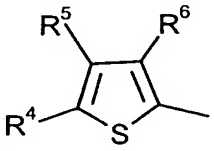
in which

R^1 represents fluorine, chlorine, bromine, iodine, methyl, isopropyl, cyclopropyl, monofluoromethyl, difluoromethyl or trifluoromethyl and

R^2 represents hydrogen, fluorine, chlorine or methyl and

R^3 represents methyl.

A furthermore very particularly preferably represents a radical of the formula

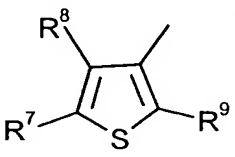


in which

R^4 and R^5 independently of one another represent hydrogen, fluorine or methyl and

R^6 represents methyl.

A furthermore very particularly preferably represents a radical of the formula

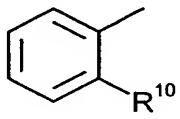


in which

R^7 and R^8 independently of one another represent hydrogen, fluorine or methyl and

R^9 represents methyl.

A furthermore very particularly preferably represents a radical of the formula

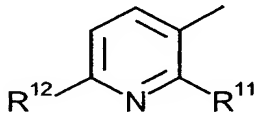


in which

R^{10} represents iodine, methyl, difluoromethyl or trifluoromethyl,

R^{10} furthermore represents chlorine or bromine.

A furthermore very particularly preferably represents a radical of the formula

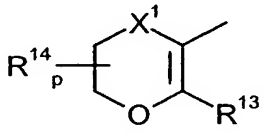


in which

R^{11} represents fluorine, chlorine, bromine, methyl or trifluoromethyl and

R^{12} represents hydrogen.

A furthermore very particularly preferably represents a radical of the formula



in which

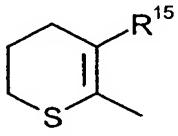
R^{13} represents methyl or trifluoromethyl and

R^{14} represents methyl,

X^1 represents S (sulfur) or CH_2 and

p represents 0 or 1.

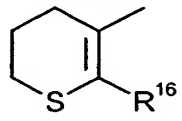
A furthermore very particularly preferably represents a radical of the formula



in which

R^{15} represents methyl, trifluoromethyl or difluoromethyl.

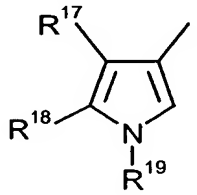
A furthermore very particularly preferably represents a radical of the formula



in which

R^{16} represents methyl, trifluoromethyl or difluoromethyl.

A furthermore very particularly preferably represents a radical of the formula



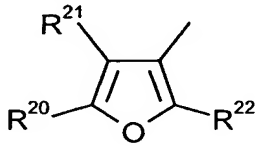
in which

R^{17} represents fluorine, methyl or trifluoromethyl,

R^{18} represents hydrogen or methyl and

R^{19} represents methyl or methoxymethyl.

A furthermore very particularly preferably represents a radical of the formula

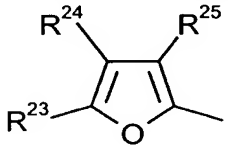


in which

R^{20} and R^{21} independently of one another represent hydrogen, fluorine or methyl and

R^{22} represents methyl or trifluoromethyl.

A furthermore very particularly preferably represents a radical of the formula

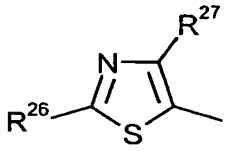


in which

R^{23} and R^{24} independently of one another represent hydrogen, fluorine or methyl and

R^{25} represents methyl.

A furthermore very particularly preferably represents a radical of the formula

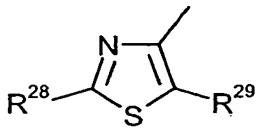


in which

R^{26} represents fluorine, chlorine, amino or methyl and

R^{27} represents chlorine, methyl, trifluoromethyl or difluoromethyl.

A furthermore very particularly preferably represents a radical of the formula

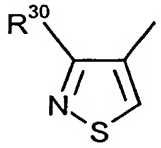


in which

R^{28} represents fluorine, chlorine, amino or methyl and

R^{29} represents chlorine, methyl, trifluoromethyl or difluoromethyl.

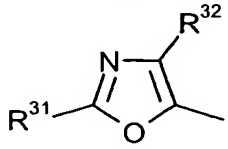
A furthermore very particularly preferably represents a radical of the formula



in which

R^{30} represents chlorine, methyl, trifluoromethyl or difluoromethyl.

A furthermore very particularly preferably represents a radical of the formula

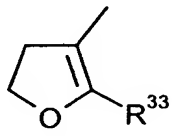


in which

R^{31} represents methyl and

R^{32} represents chlorine or methyl.

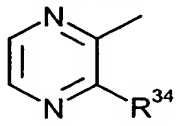
A furthermore very particularly preferably represents a radical of the formula



in which

R^{33} represents methyl, trifluoromethyl or difluoromethyl.

A furthermore very particularly preferably represents a radical of the formula



in which

R^{34} represents hydrogen, chlorine or methyl.

R especially preferably represents hydrogen or methyl.

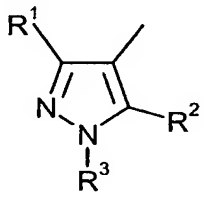
Z especially preferably represents allyl, 2-methyl-allyl, 1-methyl-allyl, 3-methyl-2-butenyl, propargyl, 2-butynyl, 3,3-difluoroallyl, cyclopropylmethyl.

X especially preferably represents fluorine.

m especially preferably represents 0 or 1.

n especially preferably represents 0.

A especially preferably represents a radical of the formula



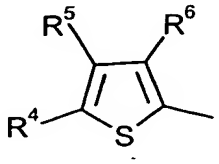
in which

R^1 represents methyl, monofluoromethyl, difluoromethyl or trifluoromethyl and

R^2 represents hydrogen, fluorine, chlorine or methyl and

R^3 represents methyl.

A furthermore especially preferably represents a radical of the formula

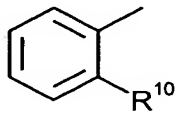


in which

R^4 and R^5 each represent hydrogen and

R^6 represents methyl.

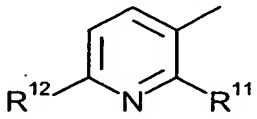
A furthermore especially preferably represents a radical of the formula



in which

R^{10} represents iodine, methyl or trifluoromethyl.

A furthermore especially preferably represents a radical of the formula

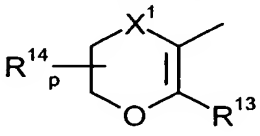


in which

R^{11} represents chlorine or trifluoromethyl and

R^{12} represents hydrogen.

A furthermore especially preferably represents a radical of the formula



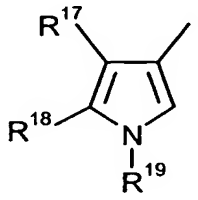
in which

R^{13} represents methyl or trifluoromethyl and

X^1 represents S (sulfur) and

p represents 0.

A furthermore especially preferably represents a radical of the formula



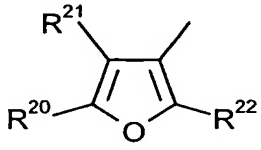
in which

R^{17} represents methyl or trifluoromethyl,

R^{18} represents hydrogen or methyl and

R^{19} represents methyl.

A furthermore especially preferably represents a radical of the formula

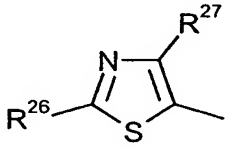


in which

R^{20} and R^{21} each represent hydrogen and

R^{22} represents methyl.

A furthermore especially preferably represents a radical of the formula

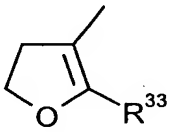


in which

R^{26} represents amino or methyl and

R^{27} represents methyl, trifluoromethyl or difluoromethyl.

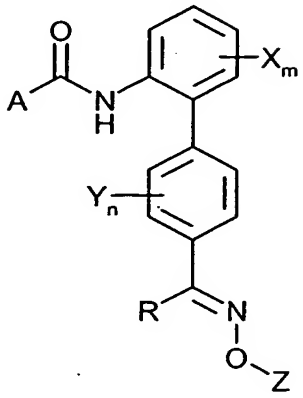
A furthermore especially preferably represents a radical of the formula



in which

R^{33} represents methyl.

Preference is furthermore given to compounds of the formula (I-1)



(I-1)

in which

R, Z, X, Y, m, n and A are as defined above. Suitable are in each case all combinations of the general, preferred, particularly preferred, very particularly preferred and especially preferred meanings mentioned above.

Particular preference is given to compounds of the formula (I-1) in which X is fluorine and m is 0 or 1.

Particular preference is given to compounds of the formula (I-1) in which n is 0.

Particular preference is given to compounds of the formula (I-1) in which R is hydrogen or methyl.

Particular preference is furthermore given to compounds of the formula (I) in which X is fluorine and m is 0 or 1.

Particular preference is furthermore given to compounds of the formula (I) in which n is 0.

Particular preference is furthermore given to compounds of the formula (I) in which R is hydrogen or methyl.

Preferred, particularly preferred, very particularly preferred and especially preferred are compounds which carry the substituents mentioned under preferred, particularly preferred, very particularly preferred and especially preferred, respectively.

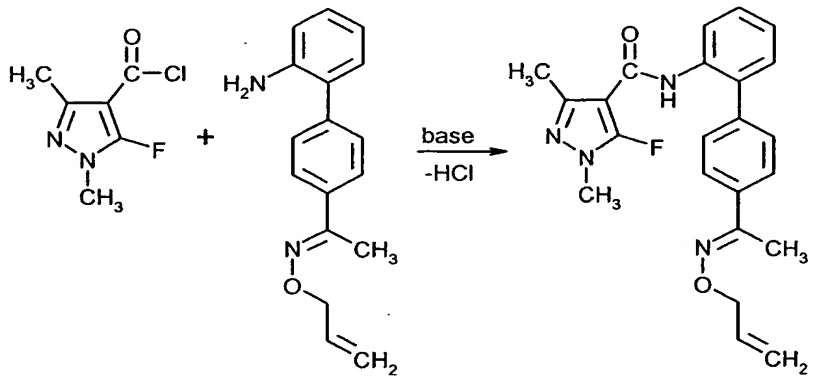
Saturated or unsaturated hydrocarbon radicals, such as alkyl or alkenyl, can in each case be straight-chain or branched as far as this is possible, including in combination with heteroatoms, such as, for example, in alkoxy.

Optionally substituted radicals can be mono- or polysubstituted, where in the case of polysubstitution the substituents can be identical or different. A plurality of radicals having the same indices, such as, for example, m radicals X for $m > 1$, can be identical or different.

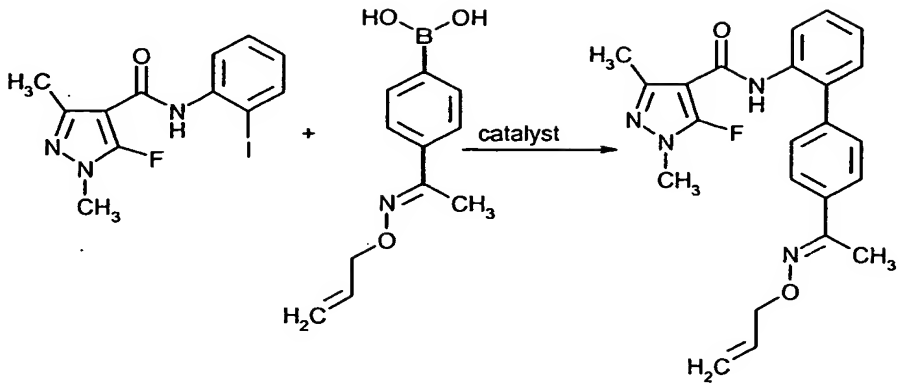
Halogen-substituted radicals, such as, for example, haloalkyl, are mono- or polyhalogenated. In the case of polyhalogenation, the halogen atoms can be identical or different. Here, halogen represents fluorine, chlorine, bromine and iodine, in particular fluorine, chlorine and bromine.

However, the general or preferred radical definitions or illustrations given above can also be combined with one another as desired, i.e. including combinations between the respective ranges and preferred ranges. The definitions apply both to the end products and, correspondingly, to precursors and intermediates. Moreover, individual definitions may not apply.

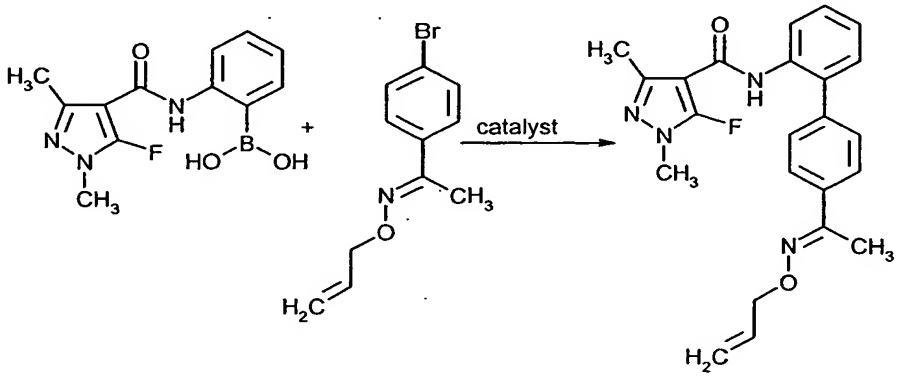
Using 5-fluoro-1,3-dimethyl-*1H*-pyrazole-4-carbonyl chloride and 1-(2'-aminobiphenyl-4-yl)ethanone *O*-allyl oxime as starting materials, the course of the process (a) according to the invention can be illustrated by the formula scheme below.



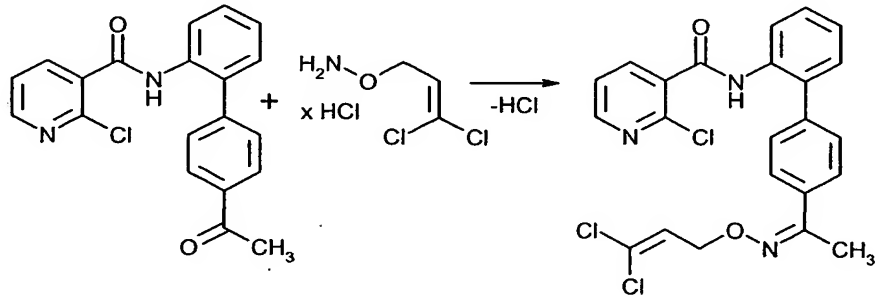
Using *N*-(2-iodophenyl)-5-fluoro-1,3-dimethyl-*1H*-pyrazole-4-carboxamide and 4-[1-(allyloxyimino)ethyl]phenylboronic acid as starting materials and a catalyst, the course of the process (b) according to the invention can be illustrated by the following scheme below.



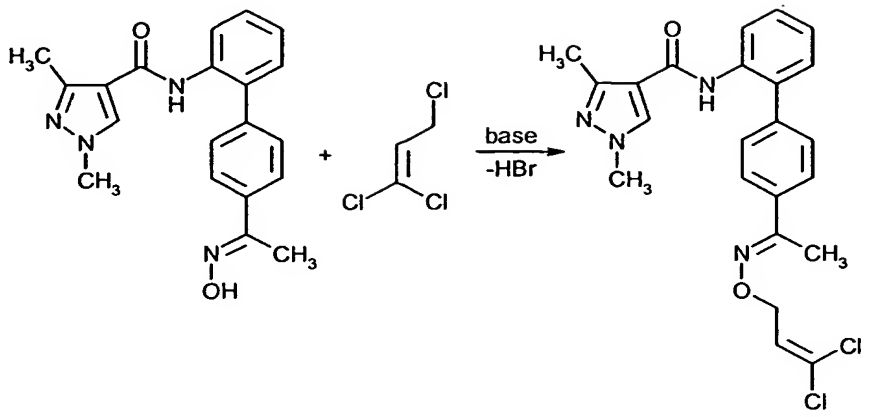
Using 2-[(5-fluoro-1,3-dimethylpyrazol-4-yl)carbonylamino]phenylboronic acid and 1-(4-bromophenyl)ethanone *O*-allyl oxime as starting materials and a catalyst, the course of the process (c) according to the invention can be illustrated by the formula scheme below.



Using *N*-(4'-acetylbiphenyl-2-yl)-2-chloronicotinamide and *O*-(3,3-dichloroallyl)hydroxylamine hydrochloride as starting materials, the course of the process (d) according to the invention can be illustrated by the formula scheme below.



Using *N*-(4'-{1-[hydroxyimino]ethyl}biphenyl-2-yl) 1,3-dimethyl-1*H*-pyrazole-4-carboxamide and 1,1,3-trichloropropene as starting materials, the course of the process (e) according to the invention can be illustrated by the formula scheme below.



Using *N*-(2-bromophenyl)-5-fluoro-1,3-dimethyl-*1H*-pyrazole-4-carboxamide and 1-(4-bromophenyl)ethanone *O*-allyl oxime as starting materials and a catalyst and 4,4,4',4',5,5,5',5'-octamethyl-2,2'-bis-1,3,2-dioxaborolane, the course of the process (f) according to the invention can be illustrated by the formula scheme below.

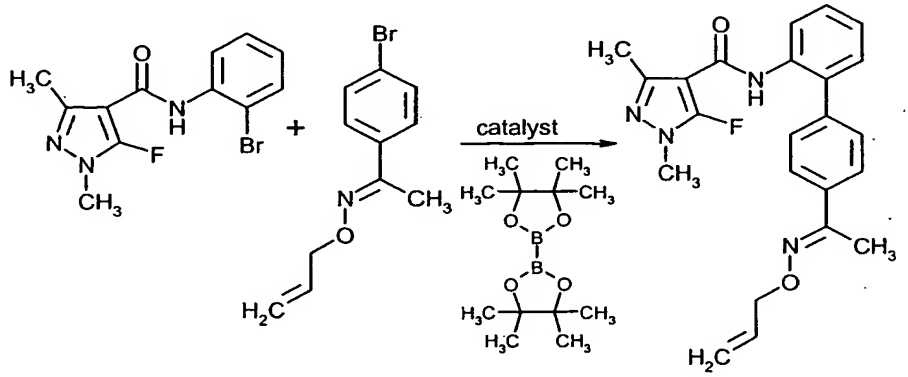


Illustration of the processes and intermediates

The formula (II) provides a general definition of the carboxylic acid derivatives required as starting materials for carrying out the process (a) according to the invention. In this formula, A preferably has those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred, very particularly preferred and especially preferred for these radicals. G preferably represents chlorine, bromine, hydroxyl, methoxy or ethoxy, particularly preferably chlorine, hydroxyl or methoxy.

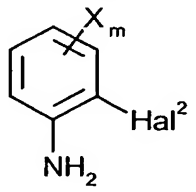
The carboxylic acid derivatives of the formula (II) are known or can be prepared by known processes (cf. WO 93/11 117, EP-A 0 545 099, EP-A 0 589 301 and EP-A 0 589 313).

The formula (III) provides a general definition of the aniline derivatives required as reaction components for carrying out the process (a) according to the invention. In this formula, R, Z, X, Y, m and n preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred, very particularly preferred and especially preferred for these radicals or these indices.

The aniline derivatives of the formula (III) are novel. Some of them can be prepared by known methods (cf. EP-A 0 545 099 and EP-A 0 589 301).

Aniline derivatives of the formula (III) are furthermore obtained when

g) 2-haloaniline derivatives of the general formula (XI)



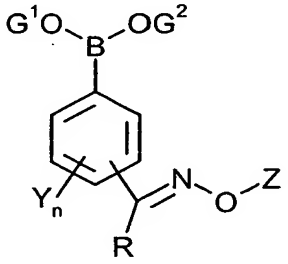
(XI)

in which

X and m are as defined above and

Hal^2 represents halogen

are reacted with boronic acid derivatives of the formula (V)

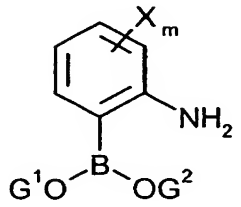


(V)

in which R, Z, Y, n, G^1 and G^2 are as defined above,

if appropriate in the presence of an acid binder, if appropriate in the presence of an inert organic diluent and if appropriate in the presence of catalyst, or

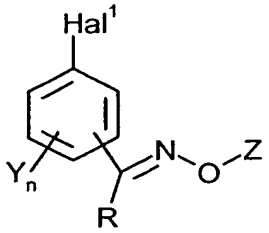
h) anilineboronic acids of the formula (XII)



(XII)

in which X, m, G^1 and G^2 are as defined bove

are reacted with phenyloxime derivatives of the formula (VII)



(VII)

in which R, Z, Y, n and Hal^1 are as defined above,

if appropriate in the presence of an acid binder, if appropriate in the presence of an inert organic diluent and if appropriate in the presence of a catalyst.

The formula (XI) provides a general definition of the 2-haloaniline derivatives required as reaction components for carrying out the processes (g) and (l) (see below) according to the invention. In this formula, X and m preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred, very particularly preferred and especially preferred for these radicals or these indices. Hal preferably represents fluorine, chlorine or bromine, particularly preferably chlorine or bromine.

The 2-haloaniline derivatives of the formula (XI) are commercially available or can be prepared from the corresponding nitro compounds by reduction.

The formula (XII) provides a general definition of the anilineboronic acids required as reaction components for carrying out the processes (h) and (j) (see below) according to the invention. In this formula, X and m preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I)

according to the invention as being preferred, particularly preferred, very particularly preferred and especially preferred for these radicals or these indices. G^1 and G^2 preferably each represent hydrogen.

Anilineboronic acids of the formula (XII) are commercially available.

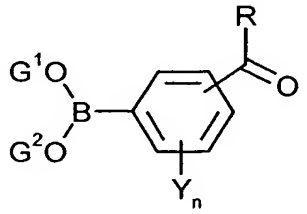
The formula (IV) provides a general definition of the carboxamide derivatives required as starting materials for carrying out the processes (b) and (f) according to the invention. In this formula, A, X and m preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred, very particularly preferred and especially preferred for these radicals or these indices.

The carboxamide derivatives of the formula (IV) are known or can be prepared by known processes (cf. WO 91/01311, EP-A 0 371 950).

The formula (V) provides a general definition of the boronic acid derivatives required for preparing the reaction components when carrying out the process (b) according to the invention and the process (g). In this formula, R, Z, Y and n preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred, very particularly preferred and especially preferred for these radicals or these indices. G^1 and G^2 preferably each represent hydrogen.

The boronic acid derivatives of the formula (V) are novel and can be prepared by

i) reacting phenylboronic acids of the formula (XIII)



(XIII)

in which R, Y, n, G^1 and G^2 are as defined above

with hydroxylamine derivatives of the formula (IX)

$$Z-O-NH_2 \times HCl \qquad (IX)$$

in which Z is as defined above,

if appropriate in the presence of an acid binder, if appropriate in the presence of an inert organic diluent and if appropriate in the presence of a catalyst.

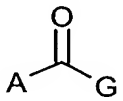
The formula (XIII) provides a general definition of the phenylboronic acids required as reaction components for carrying out the processes (i) and (l) (see below) according to the invention. In this formula, R, Y and n preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred, very particularly preferred and especially preferred for these radicals or these indices. G^1 and G^2 preferably each represent hydrogen.

The phenylboronic acids of the formula (XIII) are commercially available.

The formula (VI) provides a general definition of the carboxamide boronic acid derivatives required as reaction components for carrying out the process (c) according to the invention. In this formula, A, X and m preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred, very particularly preferred and especially preferred for these radicals or these indices. G^1 and G^2 preferably each represent hydrogen or together represent tetramethylethylene.

The carboxamide boronic acid derivatives of the formula (VI) are novel. They can be prepared by

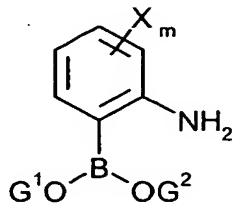
j) reacting carboxylic acid derivatives of the formula (II)



(II)

in which A and G are as defined above

with anilineboronic acids of the formula (XII)



(XII)

in which X, m, G^1 and G^2 are as defined above,

if appropriate in the presence of an acid binder, if appropriate in the presence of an inert organic diluent and if appropriate in the presence of a catalyst.

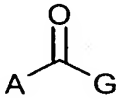
The formula (VII) provides a general definition of the phenyloxime derivatives required as reaction components for carrying out the processes (c) and (f) according to the invention and the process (h). In this formula, R, Z, Y and n preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred, very particularly preferred and especially preferred for these radicals or these indices.

The phenyloxime derivatives of the formula (VII) are known and/or can be prepared by known processes (cf. Synth. Commun. 2000, 30, 665-669, Synth. Commun. 1999, 29, 1697-1701).

The formula (VIII) provides a general definition of the biphenylacyl derivatives required as starting materials for carrying out the process (d) according to the invention. In this formula, A, R, X, Y, m and n have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred, very particularly preferred and especially preferred for these radicals or these indices.

The biphenylacyl derivatives of the formula (VIII) are novel. They can be prepared by

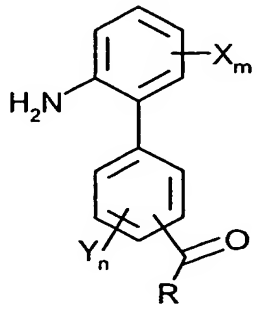
k) reacting carboxylic acid derivatives of the formula (II)



(II)

in which A and G are as defined above

with 2-benzaldehydeaniline derivatives of the formula (XIV)



(XIV)

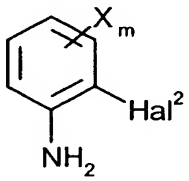
in which R, X, Y, m and n are as defined above,

if appropriate in the presence of an acid binder and if appropriate in the presence of an inert organic diluent.

The formula (XIV) provides a general definition of the 2-benzaldehydeaniline derivatives required as reaction components for carrying out the process (k) according to the invention. In this formula, R, X, Y, m and n preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred, very particularly preferred and especially preferred for these radicals or these indices.

The 2-benzaldehydeaniline derivatives of the formula (XIV) are novel. They can be prepared by

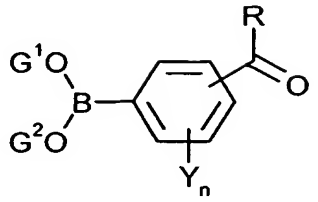
l) reacting aniline derivatives of the formula (XI)



(XI)

in which X, m and Hal^2 are as defined above and

with phenylboronic acid derivatives of the formula (XIII)



(XIII)

in which R, Y, n, G^1 and G^2 are as defined above,

if appropriate in the presence of an acid binder and if appropriate in the presence of an inert organic diluent.

The formula (IX) provides a general definition of the hydroxylamine derivatives required as reaction components for carrying out the process (d) according to the invention and the process (i). In this formula, Z preferably has those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred, very particularly preferred and especially preferred for this radical. Preference is given to using the hydrochlorides mentioned in the description. However, it is also possible to use the free hydroxylamine derivatives in the process according to the invention.

Hydroxylamine derivatives of the formula (IX) are commercially available.

The formula (I-a) provides a general definition of the hydroxyimino derivatives required as starting materials for carrying out the process (e) according to the invention. In this formula, A, R, X, Y, m and n preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred, very particularly preferred and especially preferred for these radicals or these indices.

The hydroxyimino derivatives of the formula (I-a) according to the invention can be prepared by one of the processes (a), (b), (c), (d) or (f) according to the invention described above.

The formula (X) provides a general definition of the compounds required as reaction components for carrying out the process (e) according to the invention. In this formula, Z preferably has those meanings which have already been mentioned in connection with the

description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred, very particularly preferred and especially preferred for this radical. E preferably represents chlorine, bromine, iodine, methanesulfonyl or p-toluenesulfonyl. E particularly preferably represents chlorine or bromine.

Compounds of the formula (X) are commercially available.

Suitable acid binders for carrying out the processes (a), (b), (c), (d), (e) and (f) according to the invention are in each case all inorganic and organic bases customary for such reactions. Preference is given to using alkaline earth metal or alkali metal hydroxides, such as sodium hydroxide, calcium hydroxide, potassium hydroxide, or else ammonium hydroxide, alkali metal carbonates, such as sodium carbonate, potassium carbonate, potassium bicarbonate, sodium bicarbonate, alkali metal or alkaline earth metal acetates, such as sodium acetate, potassium acetate, calcium acetate, and also tertiary amines, such as trimethylamine, triethylamine, tributylamine, N,N-dimethylaniline, pyridine, N-methylpiperidine, N,N-dimethylaminopyridine, diazabicyclooctane (DABCO), diazabicyclononene (DBN) or diazabicycloundecene (DBU). However, it is also possible to operate without added acid binder or to use an excess of the amine component so that it simultaneously acts as acid binder.

Suitable diluents for carrying out the processes (a), (b), (c), (d), (e) and (f) according to the invention are in each case all customary inert organic solvents. Preference is given to using optionally halogenated aliphatic, alicyclic or aromatic hydrocarbons, such as petroleum ether, hexane, heptane, cyclohexane, methylcyclohexane, benzene, toluene, xylene or decalin; chlorobenzene, dichlorobenzene, dichloromethane, chloroform, carbon tetrachloride, dichlorethane or trichloroethane; ethers, such as diethyl ether, diisopropyl ether, methyl t-butyl ether, methyl t-amyl ether, dioxane, tetrahydrofuran, 1,2-dimethoxyethane, 1,2-diethoxyethane or anisole; nitriles, such as acetonitrile, propionitrile, n- or isobutyronitrile or benzonitrile; amides, such as N,N-dimethylformamide, N,N-dimethylacetamide, N-methylformanilide, N-methylpyrrolidone or hexamethylphosphoric triamide; esters, such as methyl acetate or ethyl acetate, sulfoxides, such as dimethyl sulfoxide, or sulfones, such as sulfolane.

When carrying out the processes (a), (b), (c), (d), (e) and (f) according to the invention, the reaction temperatures can in each case be varied within a relatively wide range. In general, the processes are carried out at temperatures between 0°C and 140°C, preferably between 10°C and 120°C.

The process (a), (b), (c), (d), (e) and (f) according to the invention are generally in each case carried out under atmospheric pressure. However, in each case it is also possible to operate under elevated or reduced pressure.

When carrying out the process (a) according to the invention, in general 1 mol or else an excess of aniline derivative of the formula (III) and from 1 to 3 mol of acid binder are employed per mole of acid halide of the formula (II). However, it is also possible to employ the reaction components in other ratios. Work-up is carried out by customary methods. In general, water is added to the reaction mixture and the organic phase is separated off and, after drying, concentrated under reduced pressure. The residue that remains may, if required, be freed of any impurities that may still be present using customary methods, such as chromatography or recrystallization.

When carrying out the process (b) according to the invention, in general 1 mol or else an excess of boronic acid derivative of the formula (V) and from 1 to 5 mol of acid binder are employed per mole of carboxamide of the formula (IV). However, it is also possible to employ the reaction components in other ratios. Work-up is carried out by customary methods. In general, water is added to the reaction mixture and the precipitate is separated off and dried. The residue that remains may, if required, be freed of any impurities that may still be present using customary methods, such as chromatography or recrystallization.

When carrying out the process (c) according to the invention, in general 1 mol or else an excess of phenyloxime derivative of the formula (VII) and from 1 to 10 mol of acid binder and from 0.5 to 5 mol% of a catalyst are employed per mole of carboxamide boronic acid derivative of the formula (VI). However, it is also possible to employ the reaction components in other ratios. Work-up is carried out by customary methods. In general, water is added to the reaction mixture and the precipitate is separated off and dried. The residue that remains may, if required, be freed of any impurities that may still be present using customary methods, such as chromatography or recrystallization.

When carrying out the process (d) according to the invention, in general 1 mol or else an excess of hydroxylamine derivativ of the formula (IX) and from 1 to 5 mol of acid binder are employed per mole of biphenylacyl derivative of the formula (VIII). However, it is also possible to employ the reaction components in other ratios. Work-up is carried out by customary methods. In general, water is added to the reaction mixture and the precipitate is separated off, washed with water and diisopropyl ether and then dried. The residue that remains may, if required, be freed of any impurities that may still be present using customary methods, such as chromatography or recrystallization.

When carrying out the process (e) according to the invention, in general 1 mol or else an excess of reagent of the formula (X) and from 1 to 5 mol of acid binder are employed per mole of hydroxyimino derivative of the formula (I-a). However, it is also possible to employ the reaction components in other ratios. Work-up is carried out by customary methods. In general, water is added to the reaction mixture and the precipitate is separated off and dried. The residue that remains may, if required, be freed of any impurities that may still be present using customary methods, such as chromatography or recrystallization.

When carrying out the process (f) according to the invention, in general 1 mol or else an excess of phenyloxime derivative of the formula (VII) and from 1 to 5 mol of acid binder and from 1 to 5 mol of a catalyst are employed per mole of carboxamide derivative of the formula (IV). However, it is also possible to employ the reaction components in other ratios. Work-up is carried out by customary methods. In general, water is added to the reaction mixture and the precipitate is separated off and dried. The residue that remains may, if required, be freed of any impurities that may still be present using customary methods, such as chromatography or recrystallization.

The active compounds are suitable for protecting plants and plant organs, increasing harvest yields, improving the quality of the harvested material and controlling animal pests, in particular insects, arachnids and nematodes, which are encountered in agriculture, in forests, in gardens and in leisure facilities, in the protection of stored products and of materials, and in the hygiene sector and they are tolerated well by plants, have favorable homeotherm toxicity and good environmental compatibility. They may preferably be employed as crop protection agents. They are active against normally sensitive and

resistant species and against all or some stages of development. The abovementioned pests include:

From the order of the Isopoda, for example, Oniscus asellus, Armadillidium vulgare and Porcellio scaber.

From the order of the Diplopoda, for example, Blaniulus guttulatus.

From the order of the Chilopoda, for example, Geophilus carpophagus and Scutigera spp.

From the order of the Symphyla, for example, Scutigerella immaculata.

From the order of the Thysanura, for example, Lepisma saccharina.

From the order of the Collembola, for example, Onychiurus armatus.

From the order of the Orthoptera, for example, Acheta domesticus, Gryllotalpa spp., Locusta migratoria migratorioides, Melanoplus spp. and Schistocerca gregaria.

From the order of the Blattaria, for example, Blatta orientalis, Periplaneta americana, Leucophaea maderae and Blattella germanica.

From the order of the Dermaptera, for example, Forficula auricularia.

From the order of the Isoptera, for example, Reticulitermes spp.

From the order of the Phthiraptera, for example, Pediculus humanus corporis, Haematopinus spp., Linognathus spp., Trichodectes spp. and Damalinia spp.

From the order of the Thysanoptera, for example, Hercinothrips femoralis, Thrips tabaci, Thrips palmi and Frankliniella accidentalis.

From the order of the Heteroptera, for example, Eurygaster spp., Dysdercus intermedius, Piesma quadrata, Cimex lectularius, Rhodnius prolixus and Triatoma spp.

From the order of the Homoptera, for example, Aleurodes brassicae, Bemisia tabaci, Trialeurodes vaporariorum, Aphis gossypii, Brevicoryne brassicae, Cryptomyzus ribis, Aphis fabae, Aphis pomi, Eriosoma lanigerum, Hyalopterus arundinis, Phylloxera vastatrix, Pemphigus spp., Macrosiphum avenae, Myzus spp., Phorodon humuli, Rhopalosiphum padi, Empoasca spp., Euscelis bilobatus, Nephotettix cincticeps, Lecanium corni, Saissetia oleae, Laodelphax striatellus, Nilaparvata lugens, Aonidiella aurantii, Aspidiotus hederae, Pseudococcus spp. and Psylla spp.

From the order of the Lepidoptera, for example, Pectinophora gossypiella, Bupalus piniarius, Cheimatobia brumata, Lithocolletis blancardella, Hyponomeuta padella, Plutella xylostella, Malacosoma neustria, Euproctis chrysorrhoea, Lymantria spp., Bucculatrix thurberiella, Phyllocnistis citrella, Agrotis spp., Euxoa spp., Feltia spp., Earias insulana,

Heliothis spp., Mamestra brassicae, Panolis flammea, Spodoptera spp., Trichoplusia ni, Carpocapsa pomonella, Pieris spp., Chilo spp., Pyrausta nubilalis, Ephestia kuehniella, Galleria mellonella, Tineola bisselliella, Tinea pellionella, Hofmannophila pseudospretella, Cacoecia podana, Capua reticulana, Choristoneura fumiferana, Clysia ambiguella, Homona magnanima, Tortrix viridana, Cnaphalocerus spp. and Oulema oryzae.

From the order of the Coleoptera, for example, Anobium punctatum, Rhizopertha dominica, Bruchidius obtectus, Acanthoscelides obtectus, Hylotrupes bajulus, Agelastica alni, Leptinotarsa decemlineata, Phaedon cochleariae, Diabrotica spp., Psylliodes chrysocephala, Epilachna varivestis, Atomaria spp., Oryzaephilus surinamensis, Anthonomus spp., Sitophilus spp., Otiorrhynchus sulcatus, Cosmopolites sordidus, Ceuthorrhynchus assimilis, Hypera postica, Dermestes spp., Trogoderma spp., Anthrenus spp., Attagenus spp., Lyctus spp., Meligethes aeneus, Ptinus spp., Niptus hololeucus, Gibbium psylloides, Tribolium spp., Tenebrio molitor, Agriotes spp., Conoderus spp., Melolontha melolontha, Amphimallon solstitialis, Costelytra zealandica and Lissorhoptrus oryzophilus.

From the order of the Hymenoptera, for example, Diprion spp., Hoplocampa spp., Lasius spp., Monomorium pharaonis and Vespa spp.

From the order of the Diptera, for example, Aedes spp., Anopheles spp., Culex spp., Drosophila melanogaster, Musca spp., Fannia spp., Calliphora erythrocephala, Lucilia spp., Chrysomyia spp., Cuterebra spp., Gastrophilus spp., Hyppobosca spp., Stomoxys spp., Oestrus spp., Hypoderma spp., Tabanus spp., Tannia spp., Bibio hortulanus, Oscinella frit, Phorbia spp., Pegomyia hyoscyami, Ceratitis capitata, Dacus oleae, Tipula paludosa, Hylemyia spp. and Liriomyza spp.

From the order of the Siphonaptera, for example, Xenopsylla cheopis and Ceratophyllus spp.

From the class of the Arachnida, for example, Scorpio maurus, Latrodectus mactans, Acarus siro, Argas spp., Ornithodoros spp., Dermanyssus gallinae, Eriophyes ribis, Phyllocoptruta oleivora, Boophilus spp., Rhipicephalus spp., Amblyomma spp., Hyalomma spp., Ixodes spp., Psoroptes spp., Chorioptes spp., Sarcoptes spp., Tarsonemus spp., Bryobia praetiosa, Panonychus spp., Tetranychus spp., Hemitarsonemus spp. and Brevipalpus spp.

The phytoparasitic nematodes include, for example, Pratylenchus spp., Radopholus similis, Ditylenchus dipsaci, Tylenchulus semipenetrans, Heterodera spp., Globodera spp., Meloidogyne spp., Aphelenchoides spp., Longidorus spp., Xiphinema spp., Trichodorus spp., Bursaphelenchus spp.

The compounds according to the invention have potent microbicidal activity and can be employed for controlling undesirable microorganisms, such as fungi and bacteria, in crop protection and in the protection of materials.

Fungicides can be employed in crop protection for controlling Plasmodiophoromycetes, Oomycetes, Chytridiomycetes, Zygomycetes, Ascomycetes, Basidiomycetes and Deuteromycetes.

Bactericides can be employed in crop protection for controlling Pseudomonadaceae, Rhizobiaceae, Enterobacteriaceae, Corynebacteriaceae and Streptomycetaceae.

Some pathogens causing fungal and bacterial diseases which come under the generic names listed above may be mentioned as examples, but not by way of limitation:

Xanthomonas species, such as, for example, Xanthomonas campestris pv. oryzae;
Pseudomonas species, such as, for example, Pseudomonas syringae pv. lachrymans;
Erwinia species, such as, for example, Erwinia amylovora;
Pythium species, such as, for example, Pythium ultimum;
Phytophthora species, such as, for example, Phytophthora infestans;
Pseudoperonospora species, such as, for example, Pseudoperonospora humuli or Pseudoperonospora cubensis;
Plasmopara species, such as, for example, Plasmopara viticola;
Bremia species, such as, for example, Bremia lactucae;
Peronospora species, such as, for example, Peronospora pisi or P. brassicae;
Erysiphe species, such as, for example, Erysiphe graminis;
Sphaerotheca species, such as, for example, Sphaerotheca fuliginea;
Podosphaera species, such as, for example, Podosphaera leucotricha;
Venturia species, such as, for example, Venturia inaequalis;
Pyrenophora species, such as, for example, Pyrenophora teres or P. graminea

(conidia form: Drechslera, syn: Helminthosporium);
Cochliobolus species, such as, for example, Cochliobolus sativus
(conidia form: Drechslera, syn: Helminthosporium);
Uromyces species, such as, for example, Uromyces appendiculatus;
Puccinia species, such as, for example, Puccinia recondita;
Sclerotinia species, such as, for example, Sclerotinia sclerotiorum;
Tilletia species, such as, for example, Tilletia caries;
Ustilago species, such as, for example, Ustilago nuda or Ustilago avenae;
Pellicularia species, such as, for example, Pellicularia sasakii;
Pyricularia species, such as, for example, Pyricularia oryzae;
Fusarium species, such as, for example, Fusarium culmorum;
Botrytis species, such as, for example, Botrytis cinerea;
Septoria species, such as, for example, Septoria nodorum;
Leptosphaeria species, such as, for example, Leptosphaeria nodorum;
Cercospora species, such as, for example, Cercospora canescens;
Alternaria species, such as, for example, Alternaria brassicae; and
Pseudocercosporella species, such as, for example, Pseudocercosporella herpotrichoides.

The active compounds according to the invention also have very good fortifying action in plants. Accordingly, they can be used for mobilizing the defences of the plant against attack by undesirable microorganisms.

In the present context, plant-fortifying (resistance-inducing) substances are to be understood as meaning those substances which are capable of stimulating the defence system of plants such that, when the treated plants are subsequently inoculated with undesirable microorganisms, they show substantial resistance to these microorganisms.

In the present case, undesirable microorganisms are to be understood as meaning phytopathogenic fungi, bacteria and viruses. Accordingly, the substances according to the invention can be used to protect plants for a certain period after the treatment against attack by the pathogens mentioned. The period for which protection is provided generally extends over 1 to 10 days, preferably 1 to 7 days, after the treatment of the plants with the active compounds.

The fact that the active compounds are well tolerated by plants at the concentrations required for controlling plant diseases permits the treatment of above-ground parts of plants, of propagation stock and seeds, and of the soil.

The active compounds according to the invention can be used with particularly good results for controlling diseases in viticulture and in the cultivation of fruit and vegetables, such as, for example, against Venturia, Botrytis, Sclerotina, Rhizoctonia, Uncinula, Sphaerotheca, Podosphaera, Alternaria and Colletotrichum species. Rice diseases, such as Pyricularia and Pellicularia species, are likewise controlled with good results.

The active compounds according to the invention are also suitable for increasing the yield of crops. In addition, they show reduced toxicity and are well tolerated by plants.

At certain concentrations and application rates, the active compounds according to the invention can also be used as herbicides, for influencing plant growth and for controlling animal pests. If appropriate, they can also be used as intermediates and precursors for the synthesis of further active compounds.

All plants and plant parts can be treated in accordance with the invention. Plants are to be understood as meaning in the present context all plants and plant populations such as desired and undesired wild plants or crop plants (including naturally occurring crop plants). Crop plants can be plants which can be obtained by conventional plant breeding and optimization methods or by biotechnological and recombinant methods or by combinations of these methods, including the transgenic plants and inclusive of the plant cultivars protectable or not protectable by plant breeders' rights. Plant parts are to be understood as meaning all parts and organs of plants above and below the ground, such as shoot, leaf, flower and root, examples which may be mentioned being leaves, needles, stalks, stems, flowers, fruit bodies, fruits, seeds, roots, tubers and rhizomes. The plant parts also include harvested material, and vegetative and generative propagation material, for example cuttings, tubers, rhizomes, offshoots and seeds.

Treatment according to the invention of the plants and plant parts with the active compounds is carried out directly or by allowing the compounds to act on the surroundings, environment or storage space by the customary treatment methods, for

example by immersion, spraying, evaporation, fogging, scattering, painting on, injection, and, in the case of propagation material, in particular in the case of seeds, also by applying one or more coats.

In the protection of materials, the substances according to the invention can be employed for protecting industrial materials against infection with, and destruction by, undesired microorganisms.

Industrial materials in the present context are understood as meaning non-living materials which have been prepared for use in industry. For example, industrial materials which are intended to be protected by active compounds according to the invention from microbial change or destruction can be adhesives, sizes, paper and board, textiles, leather, wood, paints and plastic articles, cooling lubricants and other materials which can be infected with, or destroyed by, microorganisms. Parts of production plants, for example cooling-water circuits, which may be impaired by the proliferation of microorganisms may also be mentioned within the scope of the materials to be protected. Industrial materials which may be mentioned within the scope of the present invention are preferably adhesives, sizes, paper and board, leather, wood, paints, cooling lubricants and heat-transfer liquids, particularly preferably wood.

Microorganisms capable of degrading or changing the industrial materials which may be mentioned are, for example, bacteria, fungi, yeasts, algae and slime organisms. The active compounds according to the invention preferably act against fungi, in particular molds, wood-discoloring and wood-destroying fungi (Basidiomycetes), and against slime organisms and algae.

Microorganisms of the following genera may be mentioned as examples:
Alternaria, such as Alternaria tenuis,
Aspergillus, such as Aspergillus niger,
Chaetomium, such as Chaetomium globosum,
Coniophora, such as Coniophora puetana,
Lentinus, such as Lentinus tigrinus,
Penicillium, such as Penicillium glaucum,
Polyporus, such as Polyporus versicolor,

Aureobasidium, such as Aureobasidium pullulans,
Sclerophoma, such as Sclerophoma pityophila,
Trichoderma, such as Trichoderma viride,
Escherichia, such as Escherichia coli,
Pseudomonas, such as Pseudomonas aeruginosa, and
Staphylococcus, such as Staphylococcus aureus.

Depending on their particular physical and/or chemical properties, the active compounds can be converted to the customary formulations, such as solutions, emulsions, suspensions, powders, foams, pastes, granules, aerosols and microencapsulations in polymeric substances and in coating compositions for seeds, and ULV cool and warm fogging formulations.

These formulations are produced in a known manner, for example by mixing the active compounds with extenders, that is, liquid solvents, liquefied gases under pressure, and/or solid carriers, optionally with the use of surfactants, that is emulsifiers and/or dispersants, and/or foam formers. If the extender used is water, it is also possible to employ, for example, organic solvents as auxiliary solvents. Essentially, suitable liquid solvents are: aromatics such as xylene, toluene or alkylnaphthalenes, chlorinated aromatics or chlorinated aliphatic hydrocarbons such as chlorobenzenes, chloroethylenes or methylene chloride, aliphatic hydrocarbons such as cyclohexane or paraffins, for example petroleum fractions, alcohols such as butanol or glycol and their ethers and esters, ketones such as acetone, methyl ethyl ketone, methyl isobutyl ketone or cyclohexanone, strongly polar solvents such as dimethylformamide or dimethyl sulfoxide, or else water. Liquefied gaseous extenders or carriers are to be understood as meaning liquids which are gaseous at standard temperature and under atmospheric pressure, for example aerosol propellants such as halogenated hydrocarbons, or else butane, propane, nitrogen and carbon dioxide. Suitable solid carriers are: for example ground natural minerals such as kaolins, clays, talc, chalk, quartz, attapulgite, montmorillonite or diatomaceous earth, and ground synthetic minerals such as finely divided silica, alumina and silicates. Suitable solid carriers for granules are: for example crushed and fractionated natural rocks such as calcite, marble, pumice, sepiolite and dolomite, or else synthetic granules of inorganic and organic meals, and granules of organic material such as sawdust, coconut shells, maize cobs and tobacco stalks. Suitable emulsifiers and/or foam formers are: for example nonionic and anionic

emulsifiers, such as polyoxyethylene fatty acid esters, polyoxyethylene fatty alcohol ethers, for example alkylaryl polyglycol ethers, alkylsulphonates, alkyl sulphates, arylsulphonates, or else protein hydrolysates. Suitable dispersants are: for example lignosulphite waste liquors and methylcellulose.

Tackifiers such as carboxymethylcellulose and natural and synthetic polymers in the form of powders, granules or latices, such as gum arabic, polyvinyl alcohol and polyvinyl acetate, or else natural phospholipids such as cephalins and lecithins and synthetic phospholipids can be used in the formulations. Other possible additives are mineral and vegetable oils.

It is possible to use colorants such as inorganic pigments, for example iron oxide, titanium oxide and Prussian Blue, and organic dyestuffs such as alizarin dyestuffs, azo dyestuffs and metal phthalocyanine dyestuffs, and trace nutrients such as salts of iron, manganese, boron, copper, cobalt, molybdenum and zinc.

The formulations generally comprise between 0.1 and 95 per cent by weight of active compound, preferably between 0.5 and 90%.

The active compounds according to the invention can be used as such or in their formulations, also in a mixture with known fungicides, bactericides, acaricides, nematicides or insecticides, to broaden, for example, the activity spectrum or to prevent development of resistance. In many cases, synergistic effects are obtained, i.e. the activity of the mixture is greater than the activity of the individual components.

Examples of suitable mixing components are the following compounds:

Fungicides:

2-phenylphenol; 8-hydroxyquinoline sulfate; acibenzolar-S-methyl; aldimorph; amidoflumet; ampropylfos; ampropylfos-potassium; andoprim; anilazine; azaconazole; azoxystrobin; benalaxyl; benodanil; benomyl; benthiavalicarb-isopropyl; benzamacril; benzamacril-isobutyl; bilanafos; binapacryl; biphenyl; bitertanol; blasticidin-S; bromuconazole; bupirimate; buthiobate; butylamine; calcium polysulfide; capsimycin; captafol; captan; carbendazim; carboxin; carpropamid; carvone; chinomethionat; chlobenthiazone; chlorfenazole; chloroneb; chlorothalonil; chlozolinate; clozylacon; cyazofamid; cyflufenamid; cymoxanil;

cyproconazole; cyprodinil; cyprofuram; Dagger G; debacarb; dichlofluanid; dichlone; dichlorophen; diclocymet; diclomezine; dicloran; diethofencarb; difenoconazole; diflumetorim; dimethirimol; dimethomorph; dimoxystrobin; diniconazole; diniconazole-M; dinocap; diphenylamine; dipyrithione; ditalimfos; dithianon; dodine; drazoxolon; edifenphos; epoxiconazole; ethaboxam; ethirimol; etridiazole; famoxadone; fenamidone; fenapanil; fenarimol; fenbuconazole; fenfuram; fenhexamid; fenitropan; fenoxanil; fenpiclonil; fenpropidin; fenpropimorph; ferbam; fluazinam; flubenzimine; fludioxonil; flumetover; flumorph; fluoromide; fluoxastrobin; fluquinconazole; flurprimidol; flusilazole; flusulfamide; flutolanil; flutriafol; folpet; fosetyl-Al; fosetyl-sodium; fuberidazole; furalaxyl; furametpyr; furcarbanil; furmecyclox; guazatine; hexachlorobenzene; hexaconazole; hymexazole; imazalil; imibenconazole; iminoctadine triacetate; iminoctadine tris(albesil); iodocarb; ipconazole; iprobenfos; iprodione; iprovalicarb; irumamycin; isoprothiolane; isovaledione; kasugamycin; kresoxim-methyl; mancozeb; maneb; meferimzone; mepanipyrim; mepronil; metalaxyl; metalaxyl-M; metconazole; methasulfocarb; methfuroxam; metiram; metominostrobin; metsulfovax; mildiomycin; myclobutanil; myclozolin; natamycin; nicobifen; nitrothal-isopropyl; noviflumuron; nuarimol; ofurace; orysastrobin; oxadixyl; oxolinic acid; oxpoconazole; oxycarboxin; oxyfenthiin; paclobutrazole; pefurazoate; penconazole; pencycuron; phosdiphen; phthalide; picoxystrobin; piperalin; polyoxins; polyoxorim; probenazole; prochloraz; procymidone; propamocarb; propanosine-sodium; propiconazole; propineb; proquinazid; prothioconazole; pyraclostrobin; pyrazophos; pyrifenox; pyrimethanil; pyroquilon; pyroxyfur; pyrrolenitrine; quinconazole; quinoxyfen; quintozene; simeconazole; spiroxamine; sulfur; tebuconazole; tecloftalam; tecnazene; tetcyclacis; tetraconazole; thiabendazole; thicyofen; thifluzamide; thiophanate-methyl; thiram; tioxymid; tolclofos-methyl; tolylfluanid; triadimefon; triadimenol; triazbutil; triazoxide; tricyclamide; tricyclazole; tridemorph; trifloxystrobin; triflumizole; triforine; triticonazole; uniconazole; validamycin A; vinclozolin; zineb; ziram; zoxamide; (2S)-N-[2-[4-[[3-(4-chlorophenyl)-2-propynyl]oxy]-3-methoxyphenyl]ethyl]-3-methyl-2-[(methylsulfonyl)amino]butanamide; 1-(1-naphthalenyl)-1H-pyrrole-2,5-dione; 2,3,5,6-tetrachloro-4-(methylsulfonyl)pyridine; 2-amino-4-methyl-N-phenyl-5-thiazolecarboxamide; 2-chloro-N-(2,3-dihydro-1,1,3-trimethyl-1H-inden-4-yl)-3-pyridinecarboxamide; 3,4,5-trichloro-2,6-pyridinedicarbonitrile; actinovate; cis-1-(4-chlorophenyl)-2-(1H-1,2,4-triazol-1-yl)cycloheptanol; methyl 1-(2,3-dihydro-2,2-dimethyl-1H-inden-1-yl)-1H-imidazole-5-carboxylate; monopotassium carbonate; N-(6-methoxy-3-pyridinyl)cyclopropanecarboxamide; N-butyl-8-(1,1-dimethylethyl)-1-oxaspiro[4.5]decane-3-amine; sodium tetrathiocarbonate; and copper salts and preparations, such as Bordeaux mixture; copper hydroxide; copper naphthenate; copper oxychloride; copper sulfate; cufraneb; cuprous oxide; mancopper; oxine-copper.

Bactericides:

bronopol, dichlorophen, nitrapyrin, nickel dimethyldithiocarbamate, kasugamycin, octhilinone, furancarboxylic acid, oxytetracyclin, probenazole, streptomycin, tecloftalam, copper sulfate and other copper preparations.

Insecticides/acaricides/nematicides:

abamectin, ABG-9008, acephate, acequinocyl, acetamiprid, acetoprole, acrinathrin, AKD-1022, AKD-3059, AKD-3088, alanycarb, aldicarb, aldoxycarb, allethrin, allethrin 1R-isomers, alpha-cypermethrin (alphamethrin), amidoflumet, aminocarb, amitraz, avermectin, AZ-60541, azadirachtin, azamethiphos, azinphos-methyl, azinphos-ethyl, azocyclotin, Bacillus popilliae, Bacillus sphaericus, Bacillus subtilis, Bacillus thuringiensis, Bacillus thuringiensis strain EG-2348, Bacillus thuringiensis strain GC-91, Bacillus thuringiensis strain NCTC-11821, baculoviruses, Beauveria bassiana, Beauveria tenella, bendiocarb, benfuracarb, bensultap, benzoximate, beta-cyfluthrin, beta-cypermethrin, bifenazate, bifenthrin, binapacryl, bioallethrin, bioallethrin-S-cyclopentyl-isomer, bioethanomethrin, biopermethrin, bioresmethrin, bistrifluron, BPMC, brofenprox, bromophos-ethyl, bromopropylate, bromfenvinfos (-methyl), BTG-504, BTG-505, bufencarb, buprofezin, butathiofos, butocarboxim, butoxycarboxim, butylpyridaben, cadusafos, camphechlor, carbaryl, carbofuran, carbophenothion, carbosulfan, cartap, CGA-50439, chinomethionat, chlordane, chlordimeform, chloethocarb, chlorethoxyfos, chlorfenapyr, chlorfenvinphos, chlorfluazuron, chlormephos, chlorobenzilate, chloropicrin, chlorproxyfen, chlorpyrifos-methyl, chlorpyrifos (-ethyl), chlovaporthrin, chromafenozide, cis-cypermethrin, cis-resmethrin, cis-permethrin, clocythrin, cloethocarb, clofentezine, clothianidin, clothiazoben, codlemone, coumaphos, cyanofenphos, cyanophos, cycloprene, cycloprothrin, Cydia pomonella, cyfluthrin, cyhalothrin, cyhexatin, cypermethrin, cyphenothrin (1R-trans-isomer), cyromazine, DDT, deltamethrin, demeton-S-methyl, demeton-S-methylsulfone, diafenthiuron, dialifos, diazinon, dichlofenthion, dichlorvos, dicofol, dicrotophos, dicyclanil, diflubenzuron, dimethoate, dimethylvinphos, dinobuton, dinocap, dinotefuran, diofenolan, disulfoton, docusat-sodium, dofenapyn, DOWCO-439, eflusilanate, emamectin, emamectin-benzoate, empenthrin (1R-isomer), endosulfan, Entomopthora spp., EPN, esfenvalerate, ethiofencarb, ethiprole, ethion, ethoprophos, etofenprox, etoxazole, etrimfos, famphur, fenamiphos, fenazaquin, fenbutatin oxide, fenfluthrin, fenitrothion, fenobucarb, fenothiocarb, fenoxacrim, fenoxycarb, fenpropathrin, fenpyrad, fenpyrithrin, fenpyroximate, fensulfothion, fenthion, fentrifanil, fenvalerate, fipronil, flonicamid, fluacrypyrim, fluazuron, flubenzimine, flubrocythrinate, flucycloxuron, flucythrinate, flufenerim, flufenoxuron, flufenprox, flumethrin, flupyrazofos,

flutenzin (flufenzine), fluvalinate, fonofos, formetanate, formothion, fosmethilan, fosthiazate, fubfenprox (fluproxyfen), furathiocarb, gamma-HCH, gossyplure, grandlure, granulosis viruses, halfenprox, halofenozide, HCH, HCN-801, heptenophos, hexaflumuron, hexythiazox, hydramethylnone, hydroprene, IKA-2002, imidacloprid, imiprothrin, indoxacarb, iodofenphos, iprobenfos, isazofos, isofenphos, isoprocarb, isoxathion, ivermectin, japonilure, kadethrin, nuclear polyhedrosis viruses, kinoprene, lambda-cyhalothrin, lindane, lufenuron, malathion, mecarbam, mesulfenfos, metaldehyde, metam-sodium, methacrifos, methamidophos, Metharhizium anisopliae, Metharhizium flavoviride, methidathion, methiocarb, methomyl, methoprene, methoxychlor, methoxyfenozide, metolcarb, metoxadiazone, mevinphos, milbemectin, milbemycin, MKI-245, MON-45700, monocrotophos, moxidectin, MTI-800, naled, NC-104, NC-170, NC-184, NC-194, NC-196, niclosamide, nicotine, nitenpyram, nithiazine, NNI-0001, NNI-0101, NNI-0250, NNI-9768, novaluron, noviflumuron, OK-5101, OK-5201, OK-9601, OK-9602, OK-9701, OK-9802, omethoate, oxamyl, oxydemeton-methyl, Paecilomyces fumosoroseus, parathion-methyl, parathion (-ethyl), permethrin (cis-, trans-), petroleum, PH-6045, phenothrin (1R-trans isomer), phenthoate, phorate, phosalone, phosmet, phosphamidon, phosphocarb, phoxim, piperonyl butoxide, pirimicarb, pirimiphos-methyl, pirimiphos-ethyl, prallethrin, profenofos, promecarb, propaphos, propargite, propetamphos, propoxur, prothiofos, prothoate, protrifenbute, pymetrozine, pyraclofos, pyresmethrin, pyrethrum, pyridaben, pyridalyl, pyridaphenthion, pyridathion, pyrimidifen, pyriproxyfen, quinalphos, resmethrin, RH-5849, ribavirin, RU-12457, RU-15525, S-421, S-1833, salithion, sebufos, SI-0009, silafluofen, spinosad, spirodiclofen, spiromesifen, sulfluramid, sulfotep, sulprofos, SZI-121, tau-fluvalinate, tebufenozide, tebufenpyrad, tebupirimfos, teflubenzuron, tefluthrin, temephos, temivinphos, terbam, terbufos, tetrachlorvinphos, tetradifon, tetramethrin, tetramethrin (1R-isomer), tetrasul, theta-cypermethrin, thiacloprid, thiamethoxam, thiapronil, thiatriphos, thiocyclam hydrogen oxalate, thiodicarb, thiofanox, thiometon, thiosultap-sodium, thuringiensin, tolfenpyrad, tralocythrin, tralomethrin, transfluthrin, triarathene, triazamate, triazophos, triazuron, trichlophenidine, trichlorfon, triflumuron, trimethacarb, vamidothion, vaniliprole, verbutin, Verticillium lecanii, WL-108477, WL-40027, YI-5201, YI-5301, YI-5302, XMC, xylylcarb, ZA-3274, zeta-cypermethrin, zolaprofos, ZXI-8901, the compound 3-methylphenyl propylcarbamate (tsumacide Z), the compound 3-(5-chloro-3-pyridinyl)-8-(2,2,2-trifluoroethyl)-8-azabicyclo[3.2.1]octane-3-carbonitrile (CAS-Reg. No. 185982-80-3) and the corresponding 3-endo-isomer (CAS-Reg. No. 185984-60-5) (cf. WO-96/37494, WO-98/25923), and also preparations which comprise insecticidally active plant extracts, nematodes, fungi or viruses.

A mixture with other known active compounds, such as herbicides, or with fertilizers and growth regulators, safeners or semicochemicals, is also possible.

In addition, the compounds of the formula (I) according to the invention also have very good antimycotic activity. They have a very broad antimycotic activity spectrum in particular against dermatophytes and yeasts, moulds and diphasic fungi (for example against Candida species, such as Candida albicans, Candida glabrata), and Epidermophyton floccosum, Aspergillus species, such as Aspergillus niger and Aspergillus fumigatus, Trichophyton species, such as Trichophyton mentagrophytes, Microsporon species such as Microsporon canis and audouinii. The list of these fungi by no means limits the mycotic spectrum covered, but is only for illustration.

The active compounds can be used as such, in the form of their formulations or the use forms prepared therefrom, such as ready-to-use solutions, suspensions, wettable powders, pastes, soluble powders, dusts and granules. Application is carried out in a customary manner, for example by watering, spraying, atomizing, broadcasting, dusting, foaming, spreading, etc. It is furthermore possible to apply the active compounds by the ultra-low-volume method, or to inject the active compound preparation or the active compound itself into the soil. It is also possible to treat the seeds of the plants.

When using the active compounds according to the invention as fungicides, the application rates can be varied within a relatively wide range, depending on the kind of application. For the treatment of parts of plants, the active compound application rates are generally between 0.1 and 10 000 g/ha, preferably between 10 and 1000 g/ha. For seed dressing, the active compound application rates are generally between 0.001 and 50 g per kilogram of seed, preferably between 0.01 and 10 g per kilogram of seed. For the treatment of the soil, the active compound application rates are generally between 0.1 and 10 000 g/ha, preferably between 1 and 5000 g/ha.

The compositions used for protecting industrial materials generally comprise the active compounds in an amount of from 1 to 95%, preferably from 10 to 75%.

The application concentrations of the active compounds according to the invention depend on the nature and the occurrence of the microorganisms to be controlled and on the composition of the material to be protected. The optimum amount for application can be determined by test series. In general, the application concentrations are in the range from

0.001 to 5% by weight, preferably from 0.05 to 1.0% by weight, based on the material to be protected.

The activity and the activity spectrum of the active compounds to be used according to the invention in the protection of materials, or of the compositions, concentrates or, quite generally, formulations preparable therefrom, can be increased, if appropriate, by addition of further antimicrobially active compounds, fungicides, bactericides, herbicides, insectizides or other active compounds for widening the activity spectrum or obtaining special effects such as, for example, additional protection against insects. These mixtures may have a wider activity spectrum than the compounds according to the invention.

When used as insecticides in their commercially available formulations and in the use forms prepared with these formulations, the active compounds according to the invention can furthermore be present in the form of a mixture with synergists. Synergists are compounds by which the activity of the active compounds is increased without it being necessary for the synergist added to be active itself.

When used as insecticides in their commercially available formulations and in the use forms prepared with these formulations, the active compounds according to the invention can furthermore be present in the form of a mixture with inhibitors which reduce the degradation of the active compound after application in the habitat of the plant, on the surface of parts of plants or in plant tissues.

The active compound content of the use forms prepared from the commercially available formulations can vary within broad ranges. The active compound concentration of the use forms can be from 0.0000001 up to 95% by weight of active compound, preferably between 0.0001 and 1% by weight.

They are applied in a customary manner adapted to suit the use forms.

When used against hygiene pests and pests of stored products, the active compound is distinguished by excellent residual action on wood and clay as well as good stability to alkali on limed substrates.

As already mentioned above, it is possible to treat all plants or their parts in accordance with the invention. In a preferred embodiment, wild plant species or plant varieties and plant cultivars which have been obtained by traditional biological breeding methods, such as hybridization or protoplast fusion, and the parts of these varieties and cultivars are treated. In a further preferred embodiment, transgenic plants and plant cultivars which have been obtained by recombinant methods, if appropriate in combination with conventional methods (genetically modified organisms), and their parts are treated. The term "parts" or "parts of plants" or "plant parts" has been explained above.

Plants which are treated particularly preferably in accordance with the invention are those of the plant cultivars which are in each case commercially available or in use. Plant cultivars are understood as meaning plants with new traits which have been bred either by conventional breeding, by mutagenesis or by recombinant DNA techniques. They may take the form of cultivars, biotypes and genotypes.

Depending on the plant species or plant cultivars, their location and growth conditions (soils, climate, vegetation period, nutrition), the treatment according to the invention may also result in superadditive ("synergistic") effects. Thus, for example, reduced application rates and/or a widened activity spectrum and/or an increase in the activity of the substances and compositions which can be used in accordance with the invention, better plant growth, increased tolerance to high or low temperatures, increased tolerance to drought or to salinity in the water or soil, increased flowering performance, facilitated harvesting, accelerated maturation, higher yields, higher quality and/or better nutritional value of the harvested products, better storage characteristics and/or processability of the harvested products are possible which exceed the effects which were actually to be expected.

The preferred transgenic plants or plant cultivars (those obtained by recombinant methods) to be treated in accordance with the invention include all those plants which, owing to the process of recombinant modification, were given genetic material which confers particular, advantageous, valuable traits to these plants. Examples of such properties are better plant growth, increased tolerance to high or low temperatures, increased tolerance to drought or to salinity in the water or soil, increased flowering performance, facilitated harvesting, accelerated maturation, higher yields, higher quality and/or higher nutritional value of the harvested products, better storage characteristics and/or better processability of the harvested products. Further examples of such traits, examples which must be mentioned especially, are better defence of the plants against animal and microbial pests, such as against insects, mites,

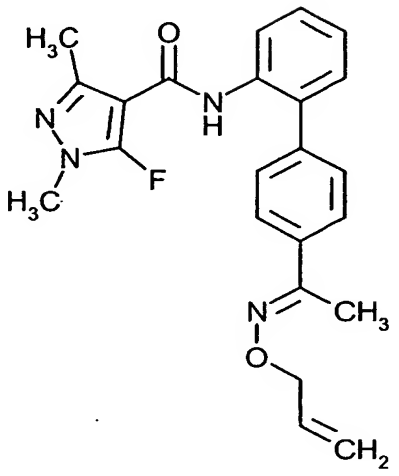
phytopathogenic fungi, bacteria and/or viruses and an increased tolerance of the plants to certain herbicidal active compounds. Examples of transgenic plants which may be mentioned are the important crop plants, such as cereals (wheat, rice), maize, soybeans, potato, cotton, tobacco, oilseed rape and fruit plants (with the fruits apples, pears, citrus fruits and grapes), with particular emphasis on maize, soybeans, potatoes, cotton, tobacco, and oilseed rape. Traits which are especially emphasized are the increased defence of the plants against insects, arachnids, nematodes, and slugs and snails owing to toxins being formed in the plants, in particular toxins which are generated in the plants by the genetic material of Bacillus thuringiensis (for example by the genes CryIA(a), CryIA(b), CryIA(c), CryIIA, CryIIIA, CryIIIB2, Cry9c Cry2Ab, Cry3Bb and CryIF and their combinations; hereinbelow "Bt plants"). Other traits which are particularly emphasized are the increased defence of plants against fungi, bacteria and viruses by the systemic acquired resistance (SAR), systemin, phytoalexins, elicitors and resistance genes and correspondingly expressed proteins and toxins. Other traits which are especially emphasized are the increased tolerance of the plants to certain herbicidal active compounds, for example imidazolinones, sulfonylureas, glyphosate or phosphinotricin (for example "PAT" gene). The genes which confer the desired traits in each case may also be present in the transgenic plants in combination with one another. Examples of "Bt plants" which may be mentioned are maize cultivars, cotton cultivars, soybean cultivars and potato cultivars which are commercially available under the trade names YIELD GARD® (for example maize, cotton, soybean), KnockOut® (for example maize), StarLink® (for example maize), Bollgard® (cotton), Nucotn® (cotton) and NewLeaf® (potato). Examples of herbicide-tolerant plants which may be mentioned are maize cultivars, cotton cultivars and soybean cultivars which are commercially available under the trade names Roundup Ready® (tolerance to glyphosate, for example maize, cotton, soybean), Liberty Link® (tolerance to phosphinotricin, for example oilseed rape), IMI® (tolerance to imidazolinones) and STS® (tolerance to sulfonylureas, for example maize). Herbicide-resistant plants (plants bred in a conventional manner for herbicide tolerance) which may be mentioned include also the varieties commercially available under the name Clearfield® (for example maize). Naturally, these statements also apply to plant cultivars having these genetic traits or genetic traits still to be developed, which plant cultivars will be developed and/or marketed in the future.

The plants listed can be treated particularly advantageously according to the invention with the compounds of the general formula (I) or the active compound mixtures according to the invention. The preferred ranges stated above for the active compounds and mixtures also apply to the treatment of these plants. Particular emphasis may be given to the treatment of plants with the compounds or mixtures specifically mentioned in the present text.

The preparation and the use of the active compounds according to the invention is illustrated by the examples below.

Preparation Examples

Example 1



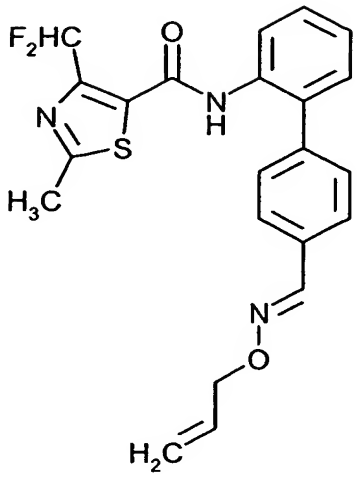
(compound 17)

Process (a)

0.300 g (1.126 mmol) of 1-(2'-aminobiphenyl-4-yl)ethanone *O*-allyl oxime and 0.114 g (1.126 mmol) of triethylamine are initially charged in 20 ml of toluene. At room temperature, 0.199 g (1.126 mmol) of 5-fluoro-1,3-dimethyl-1*H*-pyrazole-4-carbonyl chloride is added to the reaction mixture, and the mixture is stirred at 50°C for 2 h. For work-up, the reaction mixture is cooled to room temperature, washed twice with in each case 100 ml of water, dried over sodium sulfate and concentrated under reduced pressure. The residue is recrystallized from n-hexane.

This gives 0.44 g (94.4 % of theory) of N-(4'-{1-(allyloxyimino)-ethyl}-biphenyl-2-yl)-5-fluoro-1,3-dimethyl-1*H*-pyrazole-4-carboxamide (compound 17, Table 1) of melting point 118°C.

Example 2



(compound 9)

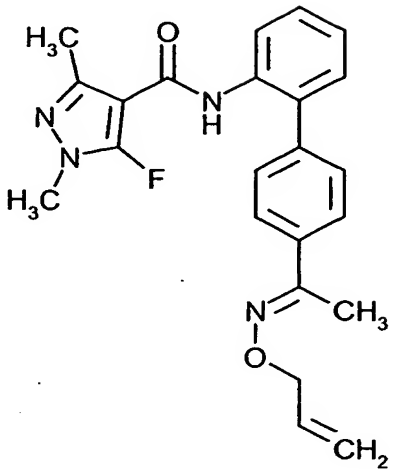
Process (a)

0.300 g (1.000 mmol) of 2'-aminobiphenyl-4-carbaldehyde *O*-allyl oxime (III-1) and 0.120 g (1.000 mmol) of triethylamine are initially charged in 15 ml of toluene. At room temperature, 0.251 g (1.000 mmol) of 4-difluoromethyl-2-methylthiazole-5-carbonyl chloride, dissolved in 5 ml of toluene, is added to the reaction mixture, and the mixture is heated at 50°C and stirred for 3 h. For work-up, the reaction mixture is cooled to room temperature and washed twice with in each case 80 ml of water, and the organic phase is dried over magnesium sulfate and then concentrated under reduced pressure. The residue is purified by silica gel column chromatography (hexane/methyl tert-butyl ether 3:1).

This gives 0.30 g (49.6% of theory) of N-[4'-(allyloxyimino-methyl)-biphenyl-2-yl]-4-difluoromethyl-2-methylthiazole-5-carboxamide (compound 9, Table 1).

^{1}H-NMR (DMSO-D_6): $\delta = 2.71$ ppm (s, 3H)

Example 3



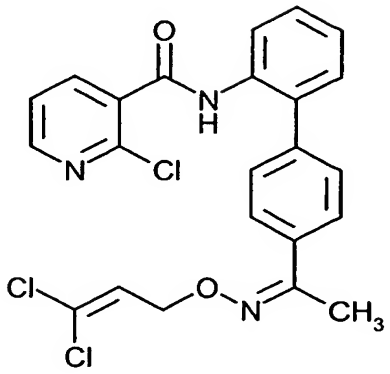
(compound 17)

Process (b)

At room temperature, 1.00 g (2.75 mmol) of *N*-(2-iodophenyl)-5-fluoro-1,3-dimethyl-*1H*-pyrazole-4-carboxamide, 0.61 g (2.78 mmol) of 4-[1-(allyloxyimino)ethyl]phenylboronic acid and 0.20 g of tetrakis(triphenylphosphine)palladium(0) are initially charged in 15 ml of dimethoxyethane. At room temperature, a solution of 1.18 g (11.14 mmol) of sodium carbonate in 15 ml of water is added with stirring. The mixture is heated at reflux temperature and stirred for 15 h. For work-up, the mixture is cooled to room temperature and extracted twice with in each case 50 ml of diethyl ether. The organic phases are washed with water, dried over sodium sulfate and concentrated under reduced pressure. The residue is purified by silica gel column chromatography (hexane/methyl tert-butyl ether 3:1).

This gives 0.13 g (10.5% of theory) of N-(4'-{1-(allyloxyimino)ethyl}biphenyl-2-yl)-5-fluoro-1,3-dimethyl-1*H*-pyrazole-4-carboxamide (compound 17, Table 1) of logP (pH 2.3) = 3.80.

Example 4



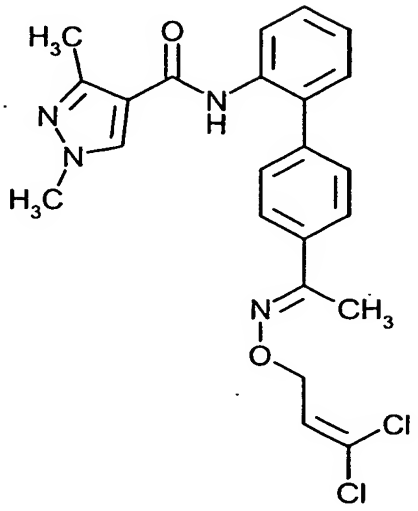
(compound 63)

Process (d)

0.79 g (2.24 mmol) of *N*-(4'-acetylbiphenyl-2-yl)-2-chloronicotinamide (VIII-1), 0.40 g (2.24 mmol) of *O*-(3,3-dichloroallyl)hydroxylamine hydrochloride and 0.22 g of sodium acetate are initially charged in a mixture of 5 ml of methanol and 2 ml of water and stirred at room temperature for 15 h. For work-up, the mixture is stirred into 30 ml of water and then extracted with 30 ml of dichloromethane. The organic phase is washed with 15 ml of water, and the phases are separated. The organic phase is dried over sodium sulfate and then concentrated under reduced pressure. The residue is triturated in a little hot hexane and cooled, and the residue is filtered off with suction.

This gives 0.68 g (63.9% of theory) of 2-chloro-*N*-{4'-[1-(3,3-dichloroallyloxyimino)ethyl]-biphenyl-2-yl}nicotinamide (compound 63, Table 1) of logP (pH 2.3) = 4.43.

Example 5



(compound 54)

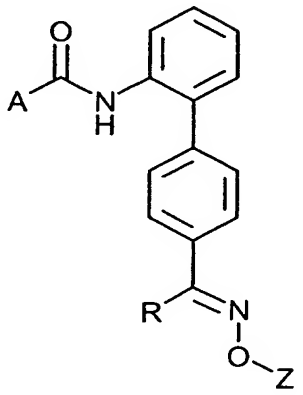
Process (e)

At room temperature, 1.00 g (2.87 mmol) of N-{4'-[1-(hydroxyimino)ethyl]biphenyl-2-yl}-1,3-dimethyl-1*H*-pyrazole-4-carboxamide and 0.60 g of potassium carbonate (4.31 mmol) are initially charged in 50 ml of acetonitrile, and 0.417 g of 1,1,3-trichloropropene (2.870 mmol), dissolved in 5 ml of acetonitrile, are then added. The mixture is heated at reflux temperature and stirred for 15 h. For work-up, the mixture is cooled and filtered off with suction through a Nutsche filter, and the mother liquor is concentrated under reduced pressure. The residue is chromatographed on silica gel (hexane/acetone 9:1).

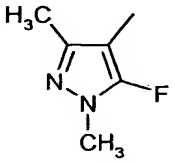
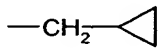
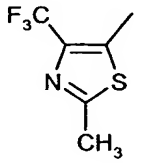
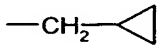
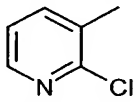
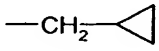
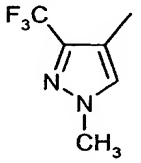
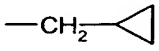
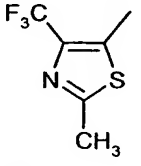
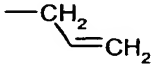
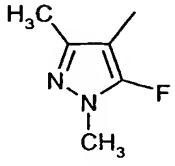
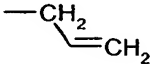
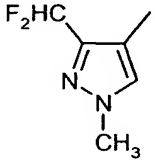
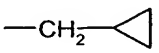
This gives 0.66 g (47.1% of theory) of N-(4'-{1-[3,3-dichloroallyloxyimino]ethyl}biphenyl-2-yl)-1,3-dimethyl-1*H*-pyrazole-4-carboxamide (compound 54, Table 1) as a solid of logP (pH 2.3) = 3.97.

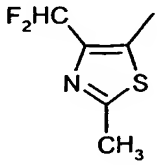
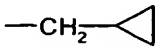
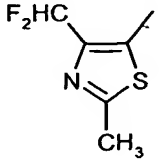
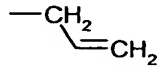
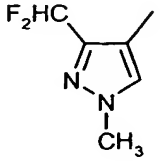
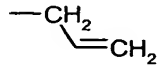
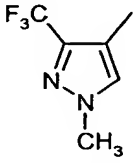
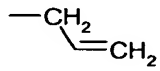
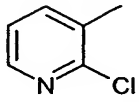
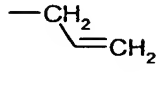
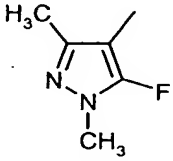
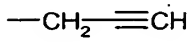
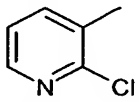
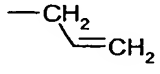
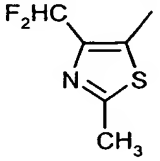
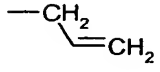
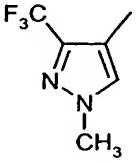
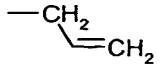
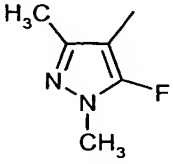
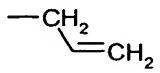
The biphenylcarboxamides of the formula (I-1) listed in the table below are likewise prepared according to Examples 1 to 5 and according to the general process descriptions and methods.

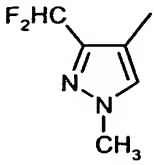
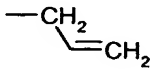
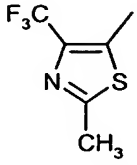
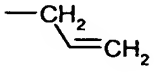
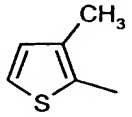
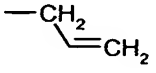
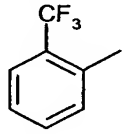
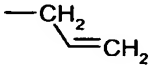
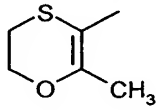
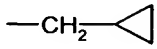
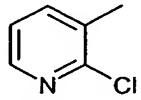
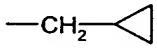
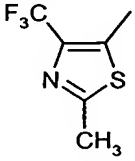
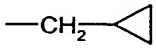
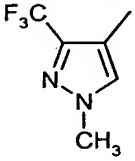
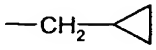
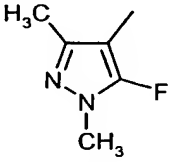
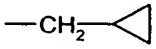
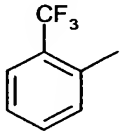
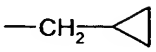
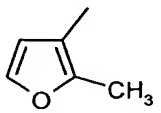
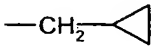
Table 1

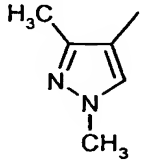
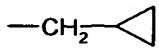
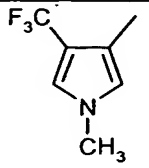
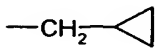
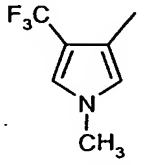
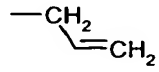
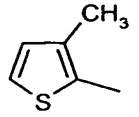
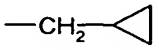
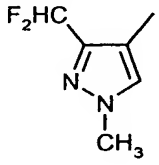
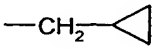
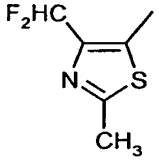
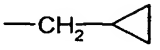
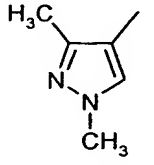
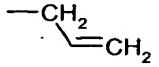
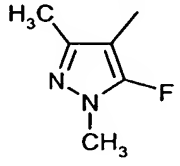
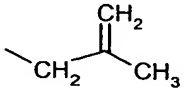
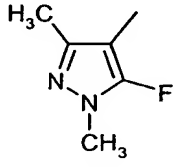
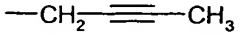
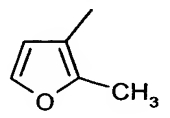
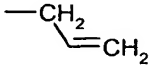


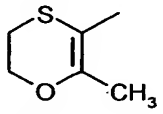
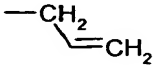
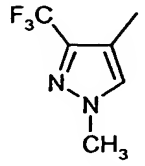
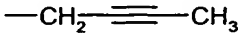
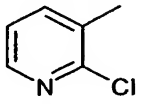
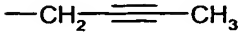
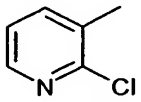
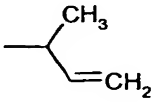
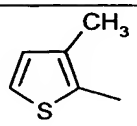
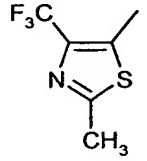
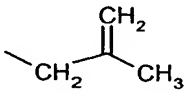
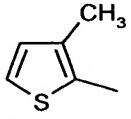
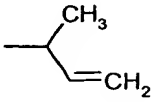
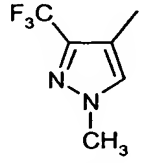
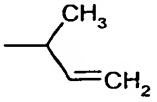
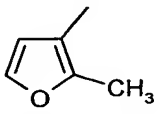
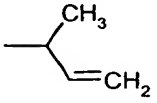
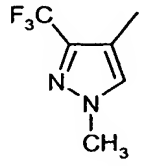
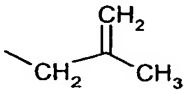
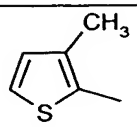
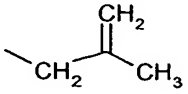
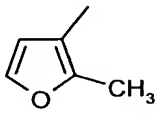
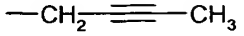
(I-1)

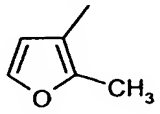
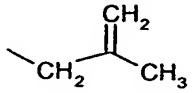
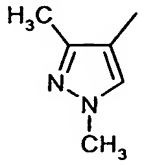
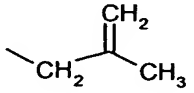
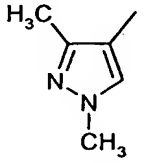
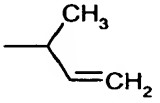
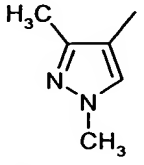
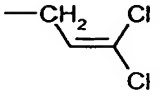
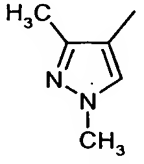
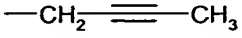
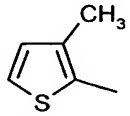
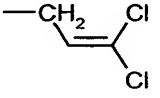
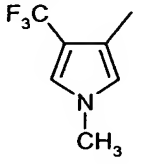
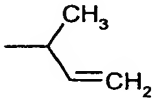
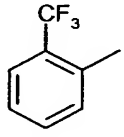
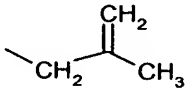
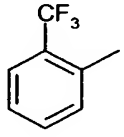
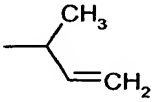
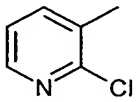
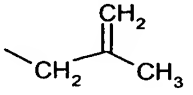
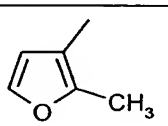
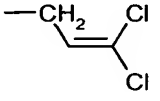
No.	A	R	Z	logP (pH 2.3)	m.p. (°C)
1		H		3.77	
2		H		4.16	
3		H		3.57	
4		H		3.77	
5		H		3.91	107-109
6		H		3.47	88-90
7		H		3.53	

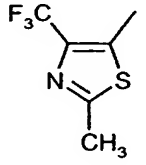
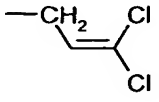
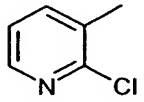
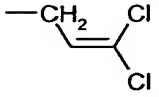
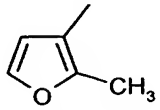
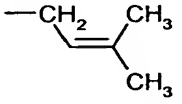
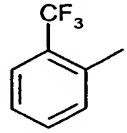
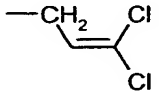
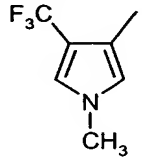
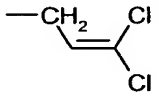
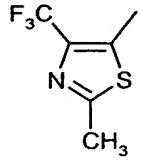
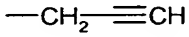
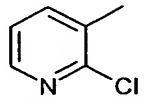
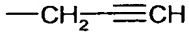
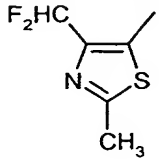
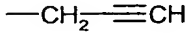
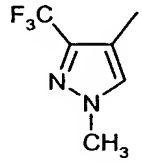
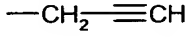
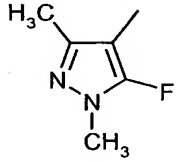
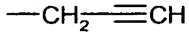
No.	A	R	Z	logP (pH 2.3)	m.p. (°C)
8		H		3.87	
9		H		3.65	
10		H		3.32	
11		H		3.53	126
12		H		3.30	88-90
13		H		3.04	
14		CH_3		3.61	78-80
15		CH_3		3.92	104-106
16		CH_3		3.81	155
17		CH_3		3.80	118

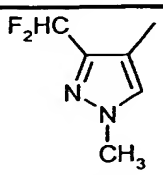
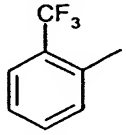
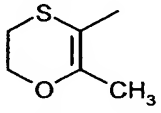
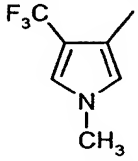
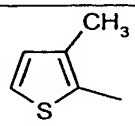
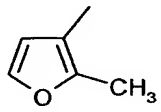
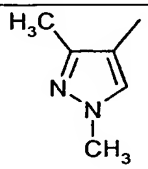
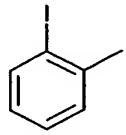
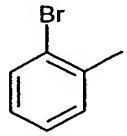
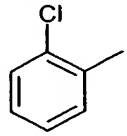
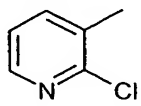
No.	A	R	Z	logP (pH 2.3)	m.p. (°C)
18		CH_3		3.57	135-137
19		CH_3		4.19	126-128
20		CH_3		4.54	76
21		CH_3		4.31	118-120
22		CH_3		4.69	78-79
23		CH_3		3.86	96-97
24		CH_3		4.45	122-124
25		CH_3		4.05	149-150
26		CH_3		4.09	141-142
27		CH_3		4.56	115-116
28		CH_3		4.43	116-117

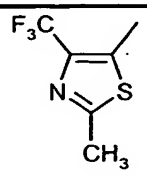
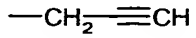
No.	A	R	Z	logP (pH 2.3)	m.p. (°C)
29		CH_3		3.42	142-144
30		CH_3		3.82	154-155
31		CH_3		4.06	152
32		CH_3		4.87	137-138
33		CH_3		3.82	170-171
34		CH_3		4.19	132-133
35		CH_3		3.16	108-110
36		CH_3		4.18	
37		CH_3		3.63	
38		CH_3		4.14	

No.	A	R	Z	logP (pH 2.3)	m.p. (°C)
39		CH_3		4.38	
40		CH_3		3.67	
41		CH_3		3.48	
42		CH_3		2.89	
43		CH_3		4.33	
44		CH_3		3.49	
45		CH_3		4,90	
46		CH_3		4.15	
47		CH_3		4.52	
48		CH_3		4.14	
49		CH_3		4.92	
50		CH_3		3.95	

No.	A	R	Z	logP (pH 2.3)	m.p. (°C)
51		CH_3		4.51	
52		CH_3		3.52	
53		CH_3		3.53	
54		CH_3		3.97	
55		CH_3		3.06	
56		CH_3		5.39	
57		CH_3		4.41	
58		CH_3		4.64	
59		CH_3		4.65	
60		CH_3		3.95	
61		CH_3		4.98	

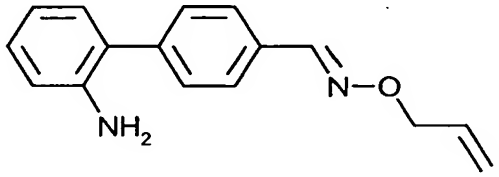
No.	A	R	Z	logP (pH 2.3)	m.p. (°C)
62		CH_3		3.81	
63		CH_3		4.43	
64		CH_3		4.84	
65		CH_3		5.05	
66		CH_3		4.83	
67		CH_3		3.72	108
68		CH_3		3.16	130-132
69		CH_3		3.45	113-115
70		CH_3		3.36	137-138
71		CH_3		3.27	109

No.	A	R	Z	logP (pH 2.3)	m.p. (°C)
72		CH_3	$-CH_2-C\equiv CH$	3.14	107
73		CH_3	$-CH_2-C\equiv CH$	3.83	140-142
74		CH_3	$-CH_2-C\equiv CH$	3.81	108-110
75		CH_3	$-CH_2-C\equiv CH$	3.6	131-133
76		CH_3	$-CH_2-C\equiv CH$	3.94	109-110
77		CH_3	$-CH_2-C\equiv CH$	3.59	
78		CH_3	$-CH_2-C\equiv CH$	2.75	98-101
79		CH_3	$-CH_2-C\equiv CH$	3.81	122
80		CH_3	$-CH_2-C\equiv CH$	3.74	116-118
81		CH_3	$-CH_2-C\equiv CH$	3.73	120
82		H	$-CH_2-C\equiv CH$	2.92	126-128

No.	A	R	Z	logP (pH 2.3)	m.p. (°C)
83		H		3.48	

Preparation of starting materials of the formula (III)

Example (III-1)



(III-1)

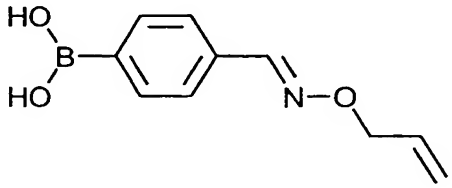
16.50 g (80.48 mmol) of 4-allyloxyiminomethylphenylboronic acid (V-1) and 13.84 g (80.48 mmol) of 2-bromoaniline together with 0.50 g (0.43 mmol) of tetrakis(triphenylphosphine)palladium are initially charged in a mixture of 100 ml of 1,2-dimethoxyethane and 100 ml of water. After addition of 34.12 g (321.92 mmol) of sodium carbonate, the mixture is heated at reflux for 12 h. For work-up, the reaction mixture is cooled to room temperature and extracted twice with in each case 200 ml of diethyl ether. The combined ether phases are washed with 400 ml of water and then dried over magnesium sulfate and concentrated under reduced pressure. The residue is chromatographed on silica gel (hexane/methyl tert-butyl ether 3:1).

This gives 4.3 g (15% of theory) of 2'-aminobiphenyl-4-carbaldehyde *O*-allyl oxime.

^{1}H-NMR (DMSO-D_6): δ = 8.33 ppm (s, 1H)

Preparation of starting materials of the formula (V)

Example (V-1)



(V-1)

In a mixture of 100 ml of methanol and 40 ml of water, 13.69 g (91.28 mmol) of formylphenylboronic acid, 10.0 g of *O*-allylhydroxylamine hydrochloride (91.28 mmol)

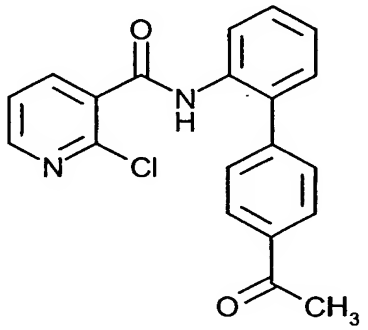
and 9.36 g (114.10 mmol) of sodium acetate are stirred at room temperature for 12 h. For work-up, the mixture is concentrated under reduced pressure and the residue is triturated with 150 ml of water, filtered off with suction through a glass frit, washed with a little water and dried on a clay disk.

This gives 16.5 g (84.6% of theory) of 4-allyloxyiminomethylphenylboronic acid.

^{1}H-NMR (DMSO-d_6): δ = 8.27 ppm (s, 1H)

Preparation of starting materials of the formula (VIII)

Example (VIII-1)



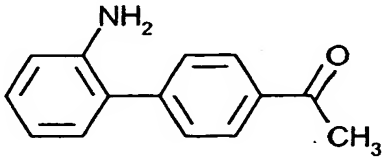
(VIII-1)

At room temperature, 8.33 g (47.33 mmol) of 2-chloronicotinoyl chloride, dissolved in 20 ml of toluene, are added over a period of 5 minutes to 10.00 g (47.33 mmol) of 1-(2'-aminobiphenyl-4-yl)ethanone (XIV-1) and 4.79 g (47.33 mmol) of triethylamine, dissolved in 150 ml of toluene. The mixture is heated at 50°C and allowed to react for 10 h. For work-up, the mixture is cooled, 100 ml of water are added and the phases are separated. The organic phase is dried over sodium sulfate and concentrated under reduced pressure. The residue is recrystallized from n-hexane.

This gives 13.7 g (82.3% of theory) of *N*-(4'-acetyl-biphenyl-2-yl)-2-chloronicotinamide of logP (pH 2.3) = 2.12.

Preparation of starting materials of the formula (XIV)

Example (XIV-1)



(XIV-1)

15.74 g (91.48 mmol) of 2-bromoaniline, 15.00 g of 4-acetylphenylboronic acid (91.48 mmol) and 0.529 g of tetrakis(triphenylphosphine)palladium(0) (0.457 mmol) are initially charged in 150 ml of dimethoxyethane. 38.78 g (365.93 mmol) of sodium carbonate, dissolved in 150 ml of water, are added over a period of 5 minutes. The mixture is heated at reflux temperature and stirred for 15 h. For work-up, the mixture is cooled and extracted twice with in each case 150 ml of ether. The combined organic phases are washed with water, dried over sodium sulfate and concentrated under reduced pressure. The residue is purified by silica gel column chromatography (cyclohexane/ethyl acetate 3:1).

This gives 15.0 g (72.6% of theory) of 1-(2'-aminobiphenyl-4-yl)ethanone of logP (pH 2.3) = 1.80.

The logP values given in Table 1 were determined with EEC Directive 79/831 Annex V.A8 by HPLC (High Performance Liquid Chromatography) on a reverse-phase column (C 18). Temperature: 43°C.

Mobile phases for the determination in the acidic range (pH 2.3): 0.1% aqueous phosphoric acid, acetonitrile; linear gradient from 10% acetonitrile to 90% acetonitrile.

Calibration was carried out using unbranched alkan-2-ones (having 3 to 16 carbon atoms) with known logP values (determination of the logP values by the retention times using linear interpolation between two successive alkanones).

The lambda-max values were determined in the maxima of the chromatographic signals using the UV spectra from 200 nm to 400 nm.

Use Examples

Example A

Venturia test (apple) / protective

Solvents:	24.5	parts by weight of acetone
	24.5	parts by weight of dimethylacetamide
Emulsifier:	1.0	part by weight of alkylaryl polyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvents and emulsifier, and the concentrate is diluted with water to the desired concentration.

To test for protective activity, young plants are sprayed with the preparation of active compound at the stated application rate. After the spray coating has dried on, the plants are inoculated with an aqueous conidia suspension of the apple scab pathogen *Venturia inaequalis* and then remain in an incubation cabin at about 20°C and 100% relative atmospheric humidity for 1 day.

The plants are then placed in a greenhouse at about 21°C and a relative atmospheric humidity of about 90%.

Evaluation is carried out 10 days after the inoculation. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

In this test, for example, the following compounds according to the invention of the Preparation Examples show good activity:

Table A:

Venturia test (apple) / protective

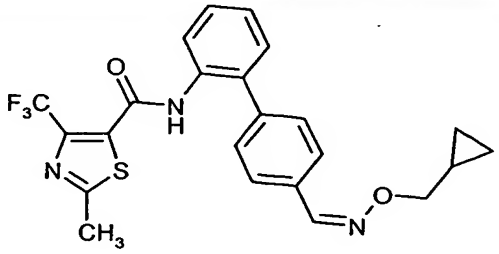
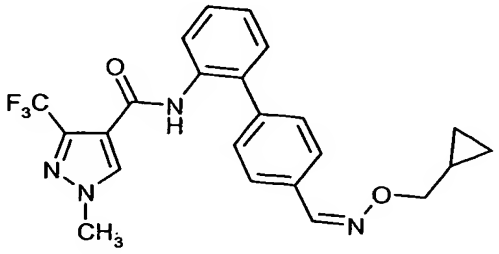
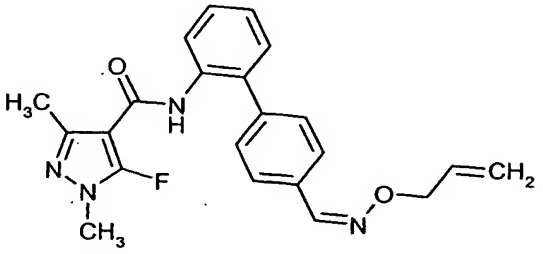
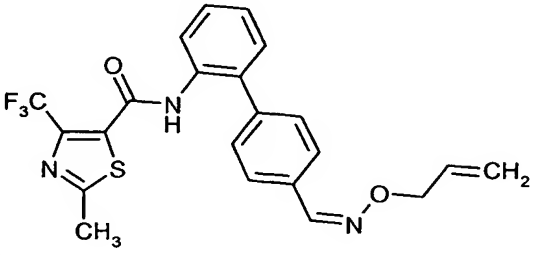
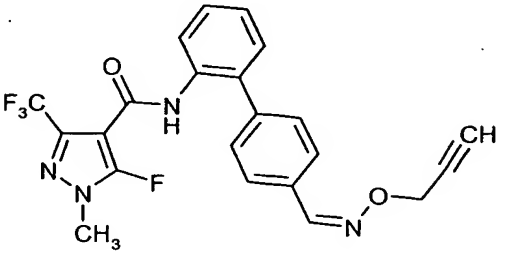
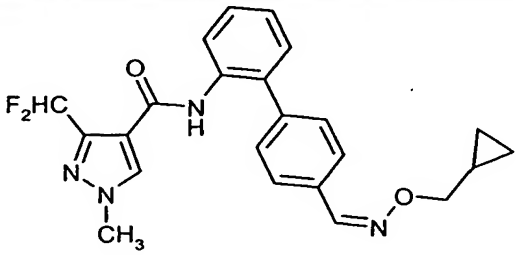
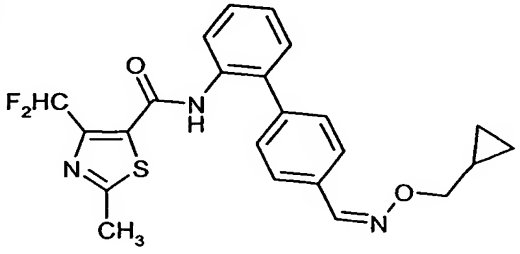
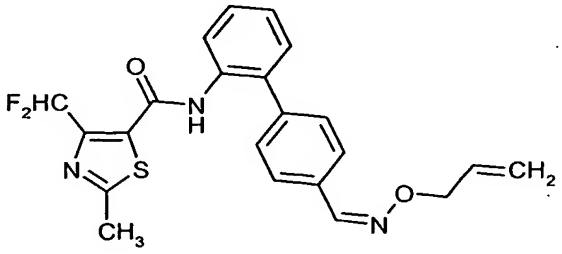
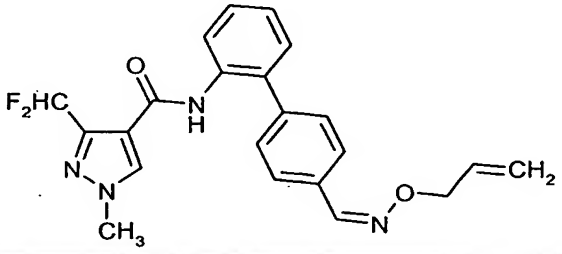
	Active compound	Active compound application rate in g/ha	Efficacy in %
2		100	98
4		100	91
6		100	97
5		100	100
13		100	100

Table A:

Venturia test (apple) / protective

Active compound		Active compound application rate in g/ha	Efficacy in %
7		100	100
8		100	100
9		100	100
10		100	100

Example B

Botrytis test (bean) / protective

Solvents:	24.5	parts by weight of acetone
	24.5	parts by weight of dimethylacetamide
Emulsifier:	1.0	part by weight of alkylaryl polyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvents and emulsifier, and the concentrate is diluted with water to the desired concentration.

To test for protective activity, young plants are sprayed with the preparation of active compound at the stated application rate. After the spray coating has dried on, 2 small pieces of agar colonized by *Botrytis cinerea* are placed onto each leaf. The inoculated plants are placed in a dark chamber at about 20°C and 100% relative atmospheric humidity.

2 days after the inoculation, the size of the infected areas on the leaves is evaluated. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

In this test, for example, the following compounds according to the invention of the Preparation Examples show good activity:

Table B:

Botrytis test (bean) / protective

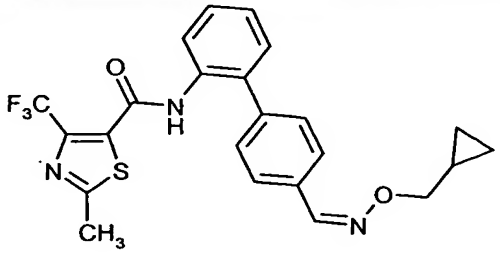
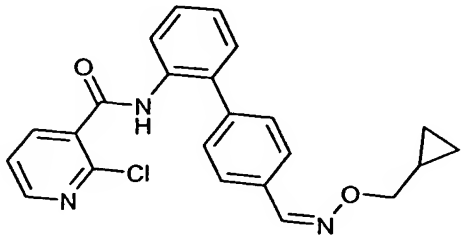
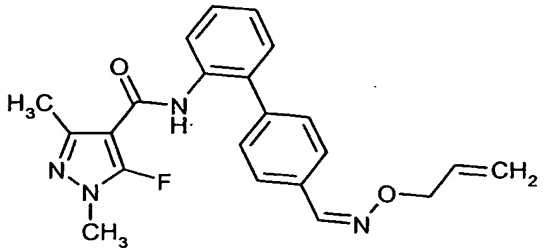
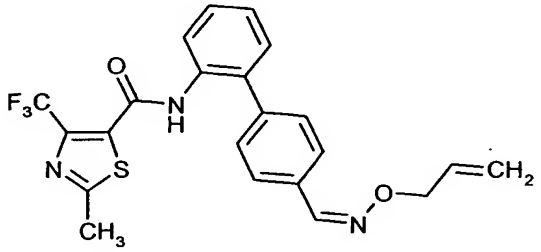
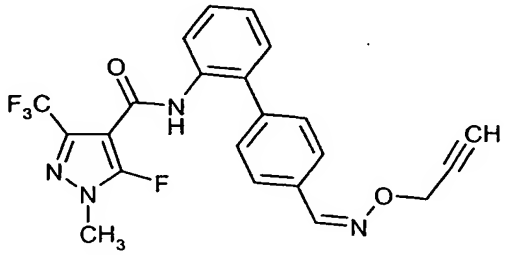
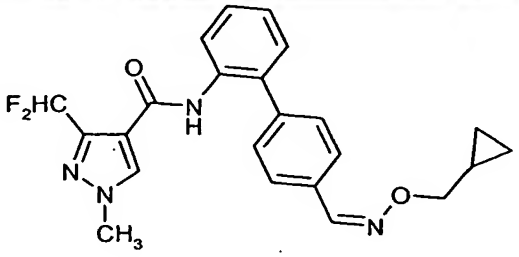
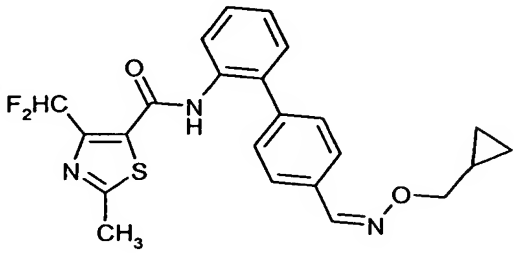
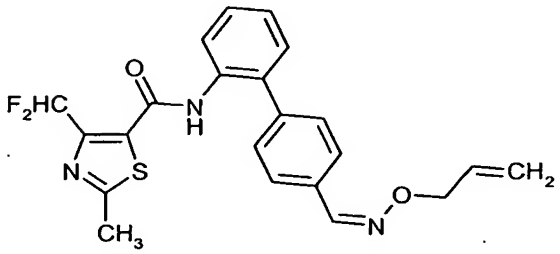
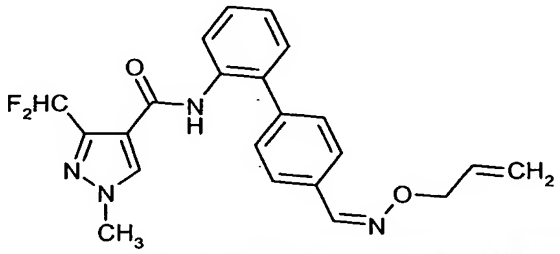
Active compound		Active compound application rate in g/ha	Efficacy in %
2		500	98
3		500	95
6		500	98
5		500	83
13		500	100

Table B:

Botrytis test (bean) / protective

Active compound		Active compound application rate in g/ha	Efficacy in %
7		500	96
8		500	85
9		500	95
10		500	95

Example C

Alternaria test (tomato) / protective

Solvent:	49	parts by weight of N,N-dimethylformamide
Emulsifier:	1	part by weight of alkylaryl polyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvent and emulsifier, and the concentrate is diluted with water to the desired concentration.

To test for protective activity, young tomato plants are sprayed with the preparation of active compound at the stated application rate. 1 day after the treatment, the plants are inoculated with a spore suspension of *Alternaria solani* and then remain at 100% relative atmospheric humidity and 20°C for 24 hours. The plants then remain at 96% relative atmospheric humidity and a temperature of 20°C.

Evaluation is carried out 7 days after the inoculation. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

In this test, for example, the following compounds according to the invention of the Preparation Examples show good activity:

Table C:

Alternaria test (tomato) / protective

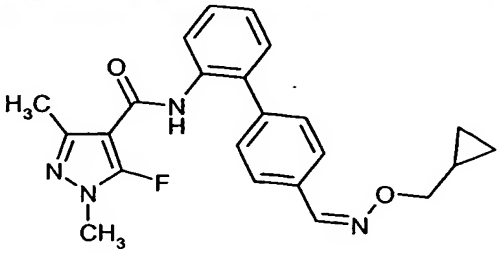
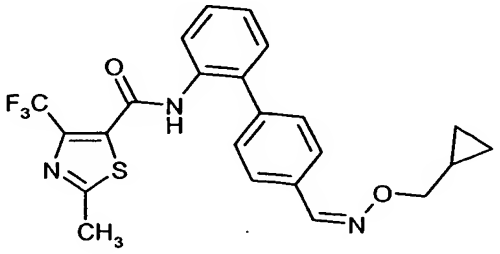
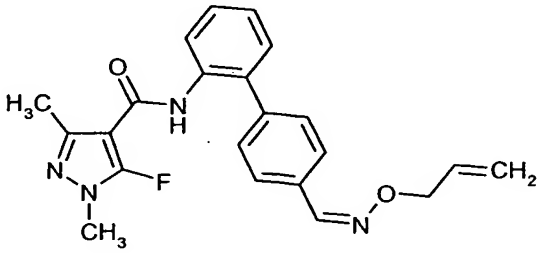
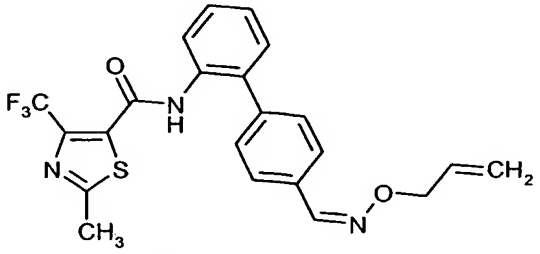
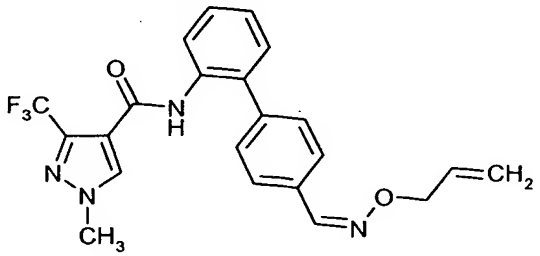
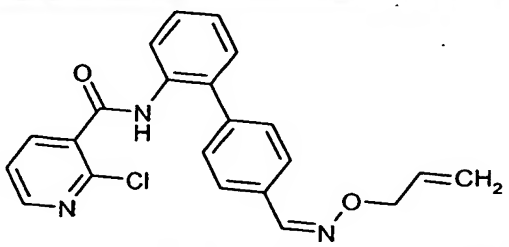
	Active compound	Active compound application rate in g/ha	Efficacy in %
1		750	90
2		750	90
6		750	100
5		750	100
11		750	100

Table C:

Alternaria test (tomato) / protective

Active compound		Active compound application rate in g/ha	Efficacy in %
12		750	100

Example D

Puccinia test (wheat) / protective

Solvents: 25 parts by weight of N,N-dimethylacetamide
Emulsifier: 0.6 parts by weight of alkylaryl polyglycol ether

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvent and emulsifier, and the concentrate is diluted with water to the desired concentration.

To test for protective activity, young plants are inoculated with a spore suspension of *Puccinia recondita* in a 0.1% strength aqueous agar solution. After the spray coating has dried on, the plants are sprayed with the preparation of active compound at the stated application rate. The plants remain in an incubation cabin at 20°C and 100% relative atmospheric humidity for 24 hours.

The plants are then placed in a greenhouse at a temperature of about 20°C and a relative atmospheric humidity of 80% to promote the development of rust pustules.

Evaluation is carried out 10 days after the inoculation. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

In this test, for example, the following compounds according to the invention of the Preparation Examples show good activity:

Table D:

Puccinia test (wheat) / protective

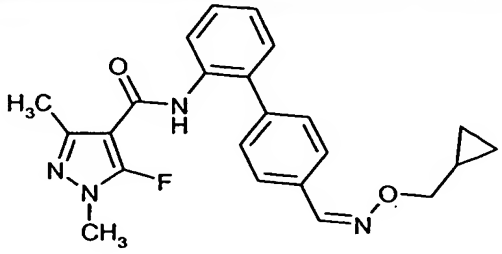
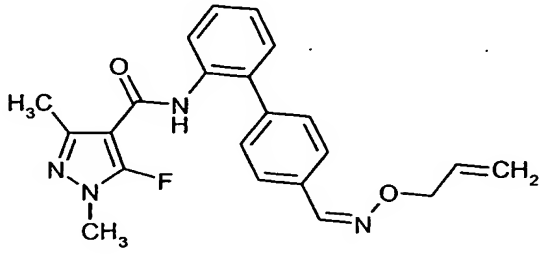
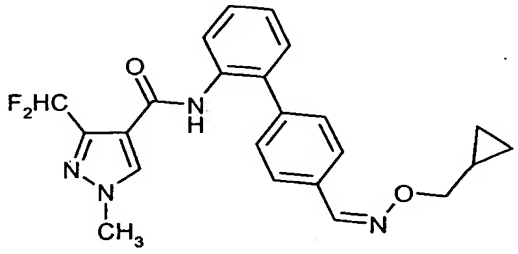
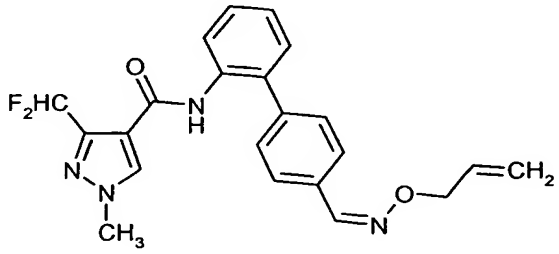
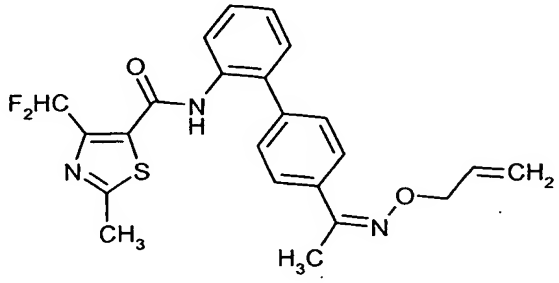
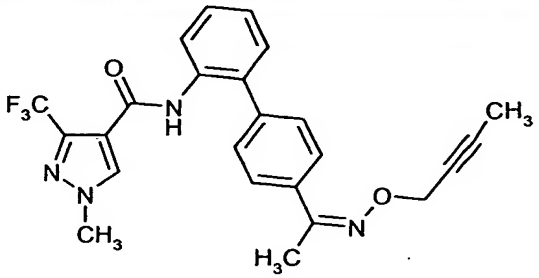
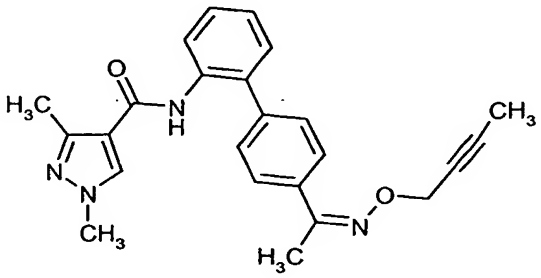
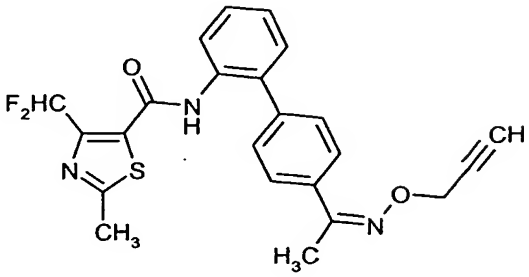
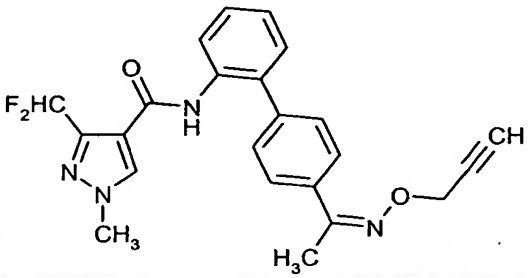
	Active compound	Active compound application rate in g/ha	Efficacy in %
1		500	100
6		500	100
7		500	100
10		500	100
15		500	100

Table D:

Puccinia test (wheat) / protective

Active compound		Active compound application rate in g/ha	Efficacy in %
40		500	100
55		500	100
69		500	100
72		500	100

Example E

Plutella test

Solvents:	100	parts by weight of acetone
	1900	parts by weight of methanol

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amount of solvent, and the concentrate is diluted with methanol to the desired concentrations.

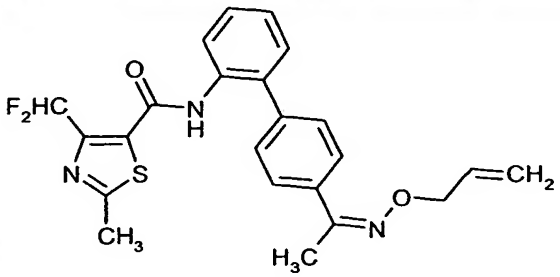
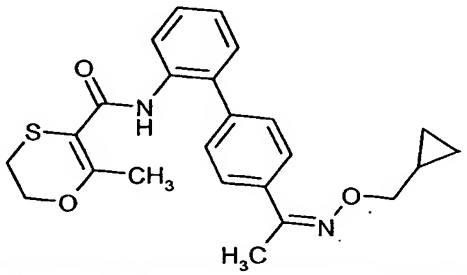
A stated amount of the preparation of active compound of the desired concentration is pipetted onto a standardized amount of synthetic feed. After the methanol has evaporated, about 200-300 eggs of the diamond back moth (*Plutella xylostella*) are placed onto the feed.

After the desired period of time, the kill of the eggs in % is determined. 100% means that all animals have been killed; 0% means that none of the animals have been killed.

In this test, for example, the following compounds of the Preparation Examples show good activity:

Table E:

Plutella test

Active compound		Active compound concentration in ppm	Kill rate in % after 7[d]
15		1000	95
22		1000	95